The Brillouin scattering of light from shear horizontal surface phonons in layered media with arbitrarily smooth interfaces

1994 J. Phys.: Condens. Matter 6 L791

(http://iopscience.iop.org/0953-8984/6/49/006)

J. Phys.: Condens. Matter 6 (1994) L791–L798. Printed in the UK

LETTER TO THE EDITOR

The Brillouin scattering of light from shear horizontal surface phonons in layered media with arbitrarily smooth interfaces

C E Bottani†‡ and R Caporali†

† Dipartimento di Ingegneria Nucleare del Politecnico di Milano, Via Ponzio 34/3, 20133 Milano, Italy

‡ INFM *Unità di Ricerca Milano Politecnico, Pza Leonardo da Vinci 32, 20133 Milano, Italy*

Received 12 October 1994

Abstract. Theoretical description of Brillouin scattering from shear horizontal (SH) acoustic surface phonons in a generic multilayer structure with arbitrarily smooth interfaces is given. To evaluate the p–s Brillouin cross-section, the SH phonon spectrum, the transmitted zeroth-order field and the fluctuating polarization vector radiating the Brillouin light are computed numerically by means of a method that can take into account any depth profiles of elastic, elasto-optic and dielectric properties of the medium including electromagnetic absorption. As an application the case of a silicon on insulator structure with both sharp and diffuse interfaces is illustrated.

Brillouin scattering of laser light has been used in the last twenty years to investigate the surface acoustic phonon spectrum of opaque and semi-opaque materials. For a review see e.g. [1]. Most of the studies have dealt with sagittal modes but a few theoretical papers [2–5] and one experimental paper [6] considered shear horizontal (SH) phonons.

More recently two papers [7, 8] have appeared treating the scattering from SH phonons in silicon on insulator (SOI) structures. These works showed and explained theoretically the existence and the dispersion properties of a surface mode localized in the buried silica layer and of a new type of pseudosurface mode quasilocalized in the top silicon layer. The examined structures had rather sharp interfaces between silicon and silica, the transition region being of the order of a few nanometres, but other SOI structures can have diffuse (smooth) interfaces between silicon and the buried silica film. Much more generally in the present paper we have in mind all the semitransparent layered structures exhibiting a gradual unidimensional (z) inhomogeneity and possessing both a strong elasto-optic coupling and a high subsurface localization of SH acoustic phonons so as to show detectable inelastic light scattering signals. The existing algorithms for computing Brillouin cross sections (p–p, p–s) in layered media assume that (i) all the material properties are space independent within each individual layer and (ii) the displacement vector field and the stress tensor are continuous across the ideally sharp interfaces connecting all neighbouring layers. Here we adopt a different and quite general approach, which is equivalent to the others in the limit of sharp interfaces. We consider the whole medium as a thick slab with two free surfaces and depth-dependent physical properties in a limited subsurface region at one side. The inhomogeneous portion of the system is described, giving the z profiles of the elastic coefficients (see figure 1), density, dielectric function and elasto-optic coefficients. All the functions of z are required to become smoothly constant in the 'bulk' portion of the system (substrate). In this way the whole phonon spectrum is discrete but becomes quasicontinuous beyond the transverse threshold of the substrate provided the slab is thick enough. In this way p–s Brillouin scattering of light from SH phonons is envisaged as a useful non-destructive spectroscopic tool to study the nature of smooth interfaces in some types of heterostructure. For the sake of simplicity the treatment is limited to cubic crystals with [001] surfaces and phonon propagation is considered only along ⟨100⟩ and ⟨110⟩ directions because of the decoupling of sagittal from SH phonon motion.

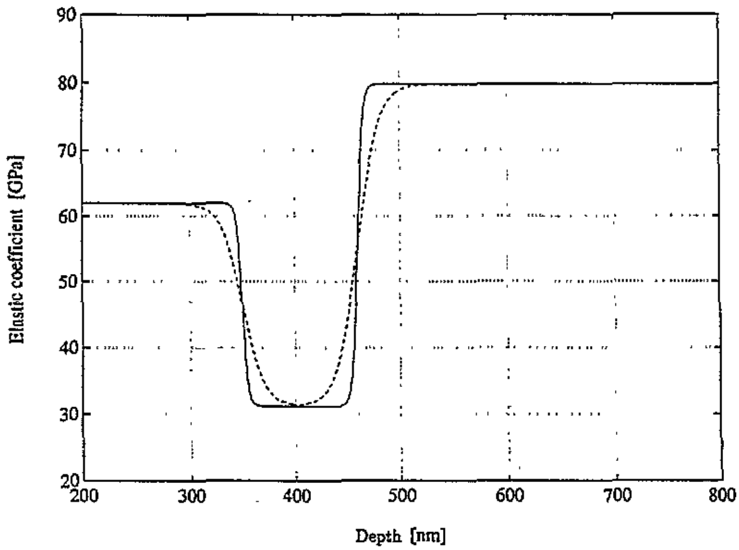


Figure 1. A typical continuous depth profile of one of the properties of the medium whose phonons scatter the light. Two interfaces are evident with two different degrees of smoothness. The medium is made up of two layers on a substrate all with different bulk elastic properties.

For the general propagation properties of elastic SH waves in layered media the reader is referred to [9]. As a consequence of the translational invariance of our system in the x direction parallel to the surface, we define the $(\omega, q_{\|})$ Fourier component of the u_y SH displacement field, being the parallel wavevector $q_{\|} = q_{\|}\hat{e}_x$, as

$$u_y(\omega, q_{\|}; x, z, t) = Q(\omega, q_{\|})\phi_y(\omega, q_{\|}, z)\exp[\mathrm{i}(q_{\|}x - \omega t)] \tag{1}$$

where $Q(\omega, q_{\|})$ is the normal coordinate of the SH phonon $(\omega, q_{\|})$. The mode z profiles $\phi_y(\omega, q_{\|}, z)$ are the real eigenfunctions, corresponding to the real eigenvalues $\omega^2 = \omega^2(q_{\|})$, of a self–adjoint Liouville equation [10]

$$(\mathrm{d}/\mathrm{d}z)\big[C_{44}(z)\ \mathrm{d}\phi_y(\omega, q_{\|}, z)/\mathrm{d}z\big] + [\rho(z)\omega^2 - C_{44}(z)q_{\|}^2]\phi_y(\omega, q_{\|}, z) = 0. \tag{2}$$

(2) is written here for the ⟨100⟩ direction. For details see [7] and [8] where the equation for the ⟨110⟩ case is also given. We assume that the illuminated surface coincides with the $z = 0$ plane and that the z axis points downwards in the medium. In the example presented below $\rho(z)$ and $C_{44}(z)$ (density and elastic constant) have a hyperbolic tangent shape at each interface, but our method is absolutely general allowing for any type of z profile. Using a slab approximation we can solve the above spectral problem in complete generality. Imposing stress–free boundary conditions at the outer surfaces and using suitable

normalization conditions [7, 8] we have always a well posed Sturm–Liouville eigenvalue problem [10].

The eigenfunctions of (2), needed to compute the p–s Brillouin cross-section (see below), can be computed numerically using the NAG routine D02KEF [11].

We assume that the free surface of the medium is illuminated by a monochromatic plane p wave with circular frequency $\omega_0 = 2\pi c/\lambda_0$. λ_0 is the corresponding wavelength in vacuum and the electric field is $(E_x, 0, E_z)$. In the absence of thermal fluctuations, cubic media would have isotropic dielectric properties. It is possible to account for the perturbation caused by long-wavelength acoustic phonons by means of an anisotropic susceptibility, that is, a second-order tensor, the components of which are linear functions of the fluctuating elastic strains [1]. Because of the linearity of the Maxwell equations and the smallness of the thermal elastic strains in thermodynamic equilibrium conditions the electric field scattered by fluctuations can be computed by first order perturbation theory (Born approximation in scattering theory) [12].

At zeroth order we compute the electric field transmitted in the medium $\mathbf{E}^{\omega_0}(x, z)$ in terms of the y component of the magnetic induction field $B_y^{\omega_0}(x, z) = e^{i(k_\parallel x)}B(z)$, where $k_\parallel = (2\pi/\lambda)\sin\theta_i = (\omega_0/c)\sin\theta_i$ is the component parallel to the surface of the wavevector of the incident wave and θ_i is the incidence angle. It is shown [13] that $B(z)$ obeys the following differential equation:

$$(\mathrm{d}/\mathrm{d}z)\big([1/\epsilon(z)]\ \mathrm{d}B/\mathrm{d}z\big) + \big(\omega_0^2/c^2 - k_\parallel^2/\epsilon(z)\big)B = 0. \tag{3}$$

$\epsilon(z)$ is the z profile of the complex relative dielectric function of the structure at frequency ω_0. In the vacuum ($z < 0$) above the surface $B(z) = (E_0/c)(e^{i(k_\perp z)} - r_p e^{-i(k_\perp z)})$, where E_0 is the electric field amplitude of the incident p wave, $k_\perp = (\omega_0/c)\cos\theta_i$ the component perpendicular to the surface of the wavevector of the incident wave and r_p the reflection coefficient. We allow for a generic z profile of a (in general) complex ϵ and impose the condition that the electromagnetic field vanish at plus infinity because of the absorbing substrate. We integrate (3) numerically using the NAG [11] FORTRAN routine D02HBF which can also evaluate r_p self-consistently. Once $B(z)$ has been computed $E_x^{\omega_0}(x, z)$ and $E_z^{\omega_0}(x, z)$ can also be obtained [13]. At first order in perturbation theory the elasto-optic coupling is described by a fluctuating polarization vector radiating the scattered light. For p–s scattering from SH phonons this has only one component $(P_y^{\omega_s})_R$ which is written [7] in terms of the fluctuating thermal elastic strains $u_{yx} = \frac{1}{2}[\partial u_y(\omega_\alpha, q_\parallel)/\partial x]$ and $u_{yz} = \frac{1}{2}[\partial u_y(\omega_\alpha, q_\parallel)/\partial z]$.

We write $(P_y^{\omega_s})_R$ in the compact form (for the sake of simplicity we report only the complex anti-Stokes term, radiating at the circular frequency $\omega_s = \omega_0 + \omega_\alpha(q_\parallel)$, corresponding to the annihilation of pre-existing phonons of the normal mode α)

$$(P_y^{\omega_s})_R^\alpha = Q(\omega_\alpha, q_\parallel)\Pi_y(z|\omega_0, k_\parallel; \omega_\alpha, q_\parallel)e^{i(k_\parallel + q_\parallel)x} \tag{4}$$

where

$$\Pi_y(z|\omega_0, k_\parallel; \omega_\alpha, q_\parallel) = \frac{c^2\epsilon_0 k_{44}(z)}{2\omega_0\epsilon(z)}\left[q_\parallel\phi_y(\omega_\alpha(q_\parallel), q_\parallel, z)\frac{\mathrm{d}B}{\mathrm{d}z} - k_\parallel\frac{\mathrm{d}\phi_y(\omega_\alpha(q_\parallel), q_\parallel, z)}{\mathrm{d}z}B(z)\right] \tag{5}$$

are spectral weights depending on both the phonon mode profiles and the zeroth-order transmitted electromagnetic field in the medium. $k_{44}(z)$ is the (generic) depth profile of the sole elasto-optic coefficient involved for ⟨100⟩ [7].

Using the found $(P_y^{\omega_s})_R^\alpha$ in the Maxwell equations, we obtain an inhomogeneous wave equation for the radiation of the scattered field component $E_y^{s\alpha}(x, z, t) = E_y^\alpha(z)e^{i(k_{\|}^s x - \omega_s t)}$ in the medium ($z > 0$), with $k_{\|}^s = k_{\|} + q_{\|}$: the rule expressing the conservation of parallel wavevector for an anti-Stokes event. This wave equation is formally identical to that used in [8] for the sharp-interface case, which now has to be integrated in the whole medium with continuously varying depth profiles. In this way the problem is reduced to the determination of $E_y^\alpha(0^-) = E_y^\alpha(0^+) = E_y^\alpha(0)$ (at the upper surface), which, propagated in vacuum at the observation point, gives the scattered amplitude. This can be accomplished, in our model, by means of numerical integration of the equation in question. $E_y^\alpha(z)$ in the medium can be found using the NAG FORTRAN routine D02HBF [11] where one has to fix the infinitesimal *initial values* (at plus infinity) of the scattered field and of its z derivative. To obtain the total fluctuating scattered field component $(\omega, \boldsymbol{k}^s)$, one has to sum over all α values (the whole SH phonon spectrum at fixed $q_{\|}$). Finally we find the differential scattering cross-section for anti-Stokes Brillouin backscattering from SH phonons as

$$\frac{d^2\sigma}{d\Omega\, d\omega} \propto \cos\theta_s \sum_\alpha \frac{|A_\alpha|^2}{\omega_\alpha^2(q_{\|})} \delta[\omega - (\omega_0 + \omega_\alpha(q_{\|})] \tag{6}$$

where the coefficients A_α are proportional to $E_y^\alpha(0)$. Detailed expressions are given elsewhere [14].

As an application we present some instances of p–s Brillouin cross-sections for scattering from SH phonons in structures consisting of a silicon/silica bilayer on an Si(001) substrate, allowing for the existence of (different) smooth interfaces between silicon and silica. This choice has been motivated as described in the introduction.

The functions describing the scattering properties of the medium have a local hyperbolic tangent profile at each interface. For example, to describe the density profile in a system consisting of a single silicon/silica bilayer on a silicon substrate we adopt a $\rho(z)$ of the type

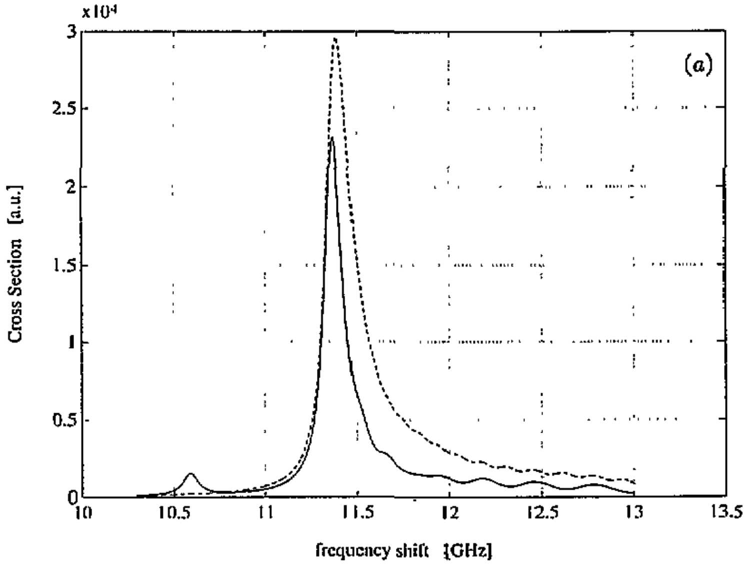
$$\rho(z) = (\rho_1 - \rho_2)\left\{1 - \frac{1}{2}\left[\tanh\left(\frac{z - d_1}{\delta_1}\right) + 1\right]\right\} + \rho_2 + (\rho_1 - \rho_2)\frac{1}{2}\left[\tanh\left(\frac{z - d_2}{\delta_2}\right) + 1\right] \tag{7}$$

where ρ_1 and ρ_2 are, respectively, the mass density of bulk silicon and silica; $d_1 = d$, $d_2 = d + L$ are, respectively, the depths of the ideally sharp first and second interfaces; and δ_1 and δ_2 characterize the degree of smoothness of the *real* interfaces. In this particular case one can imagine that the transition layers of thickness δ are constituted by SiO_x with x varying with z between zero and two.

In our computations the wavelength of the light incident onto the medium is $\lambda_0 = 514.5$ nm, typical of Ar lasers used in Brillouin spectroscopy. The physical properties of bulk silica and silicon are the same as used in [8]. The theoretical cross sections are convoluted with a Lorentzian of 200 MHz width to account for the finite experimental spectral resolution. As a general observation we stress that, while the velocity of sound is less in silica than in silicon, and so the acoustic phonons tend to get trapped within the silica layers, the elasto-optic coupling is strong in silicon but negligible in silica. Thus intense signals originate from the subsurface silicon layers where the nearby silica layers induce appreciable localization of the acoustic modes [7, 8].

Figure 2 shows the differential scattering cross sections for 30° backscattering in different silica/silicon structures.

In figure 2(*a*) the effect of the presence of an *imperfect* silicon film at the surface of a simple SOI structure is simulated reducing the k_{44} of the upper silicon by 20%. Here



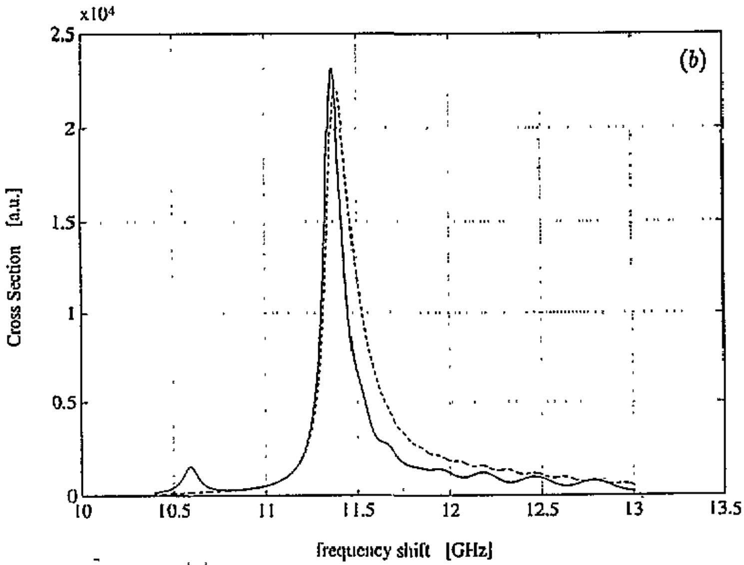


Figure 2. The theoretical cross-section at $\theta_i = 30°$ for different systems. (*a*) $\delta = 1$ nm for both interfaces. k_{44} of ideal silicon for the top layer (solid line); $k_{44} = 0.8$ of imperfect silicon for the top layer (dashed line). (*b*) $\delta_1 = \delta_2 = 1$ nm (solid line); $\delta_1 = 1$ nm and $\delta_2 = 40$ nm (dashed line). Perfect top silicon.

$d = 350$ nm and $L = 110$ nm. In this case rather sharp interfaces $\delta_1 = 1$ nm, $\delta_2 = 1$ nm are assumed. A remarkable enhancement of the peak is evident corresponding to the pseudo–Love wave, and a drastic reduction of the peak (this peak is no longer visible) corresponding to the Love wave (dashed line) with respect to the case of perfect silicon at the top (solid line).

It is possible to explain the above facts as follows. If we observe the mean square polarization radiating the Brillouin light at the frequency of the Love wave (figure 3(*a*)),

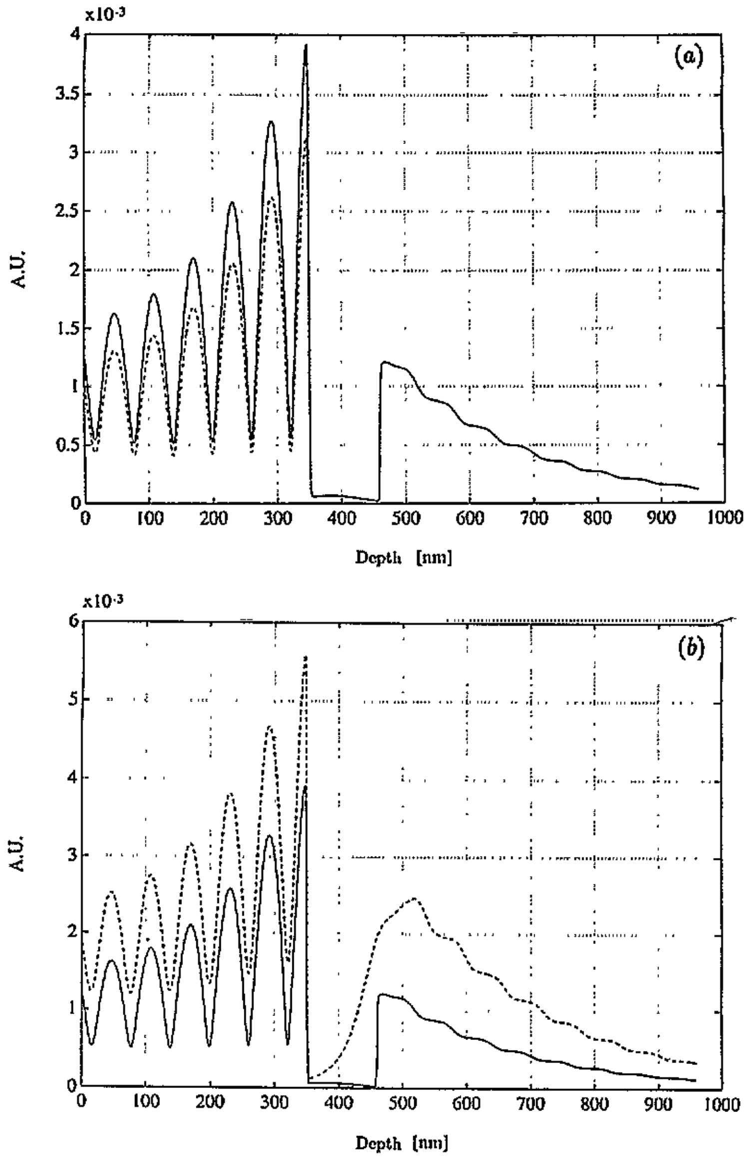


Figure 3. The mean square polarization radiating the Brillouin light against z at the frequency of different SH waves: (a) for the Love wave of figure 2(a) (first peak); perfect top silicon (solid line); imperfect top silicon (dashed line); (b) for the Love wave of figure 2(b) (first peak); with two sharp interfaces (solid line); with the second interface smooth (dashed line).

we see that it is confined in the top silicon and the silica layers and, therefore, it responds strongly to the great decrease of k_{44} in the subsurface silicon layer. We also stress that the results shown by the solid lines differ from the ideally sharp interface case presented in [8], where $\delta = 0$. In that ideal case the first peak is higher than in the present case. The differences are hidden in the electromagnetic part of the computation while the phonon spectrum does not change appreciably.

Figure 2(b) refers to the same reference structure as figure 2(a) (the same L and d) but with perfect materials. The solid line is the same as in figure 2(*a*) while the dashed line refers to the case $\delta_1 = 1$ nm, $\delta_2 = 40$ nm: that is, the second interface is imperfect (quite broad).

It can be seen that the peak corresponding to the Love wave is practically invisible, when the second interface is smeared out (dashed line), if it is compared to the case with two sharp interfaces (solid line). This is not understandable by simply analysing the corresponding polarization profiles (figure 3(*b*)) as in the previous case. Here a major role is played by the propagation of the scattered light back through the layered medium. In fact, in the case of sharp interfaces [8] this SOI structure presents a high transmissivity in the frequency band of the two SH modes, but, as the smoothness of the interfaces increases, this peculiar feature is destroyed and the light scattered by the Love mode is filtered out.

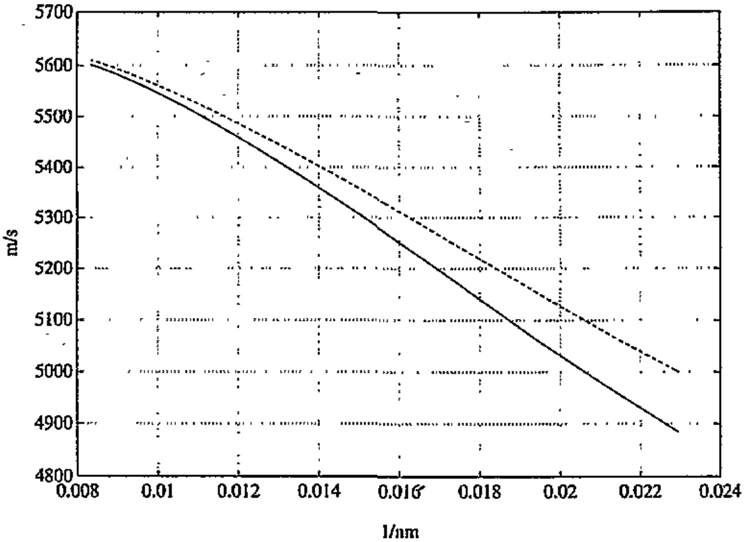


Figure 4. The dispersion relations for the Love wave in the sharp-($\delta_1 = \delta_2 = 1$ nm: solid line) and smooth-($\delta_1 = \delta_2 = 20$ nm: dashed line) interface cases.

In figure 4 we show the dispersion curves for Love waves in the sharp- (solid line) and smooth- (dashed line) interface cases. It is seen that in the latter case the phase velocity of the waves is higher. This is due to the fact that the modes are less confined in the buried silica layer and extend more in the surrounding silicon, whose sound velocity is bigger. In this case the amount of the above effect at large $q_{\|}$ values is big enough to be detected experimentally.

As a last example of the application of our method, in figure 5 we illustrate the behaviour of the modulus of the reflection coefficient as a function of the degree of smoothness of interfaces in a simple SOI structure compared to the case of semi–infinite silicon.

In conclusion, we have presented the scheme of computation of the cross-section for Brillouin scattering of light by SH surface acoustic phonons for a general layered structure with arbitrarily smooth interfaces. An example concerning imperfect silicon on insulator structures has been illustrated. We think we have demonstrated the possibility of using our numerical method for complicated systems for which even a semi-analytic study is impossible.

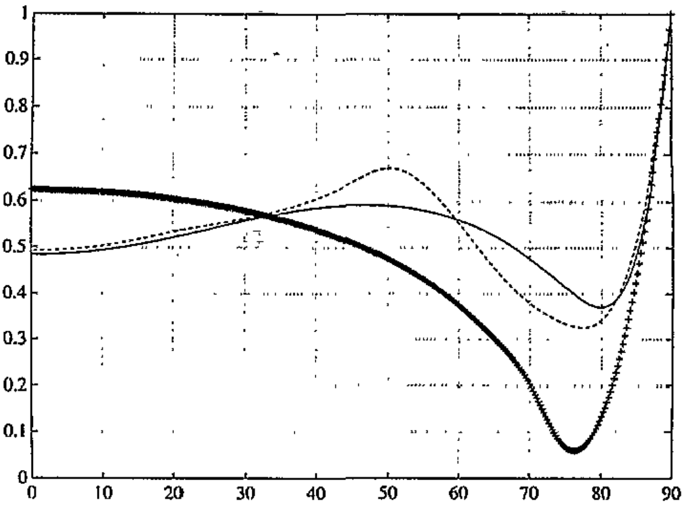


Figure 5. The modulus of the reflection coefficient r_p against incidence angle: +, semi-infinite silicon; --, the same SOI structure as in figure 2(*a*) ($\delta_1 = \delta_2 = 1$ nm); solid line, the same SOI structure as in figure 2(*a*) ($\delta_1 = \delta_2 = 5$ nm).

Many helpful discussions with Dr M Beghi, Dr G Ghislotti and Dr P Mutti are gratefully acknowledged. The presented activity was funded by progetto finalizzato *Materiali Speciali per Tecnologie Avanzate* of CNR.

References

[1] Nizzoli F and Sandercock J R 1990 *Dynamical Properties of Solids* ed G K Horton and A A Maradudin (Amsterdam: Elsevier) pp 281–335
[2] Bortolani V, Marvin A M, Nizzoli F and Santoro G 1983 *J. Phys. C: Solid State Phys.* **16** 1757
[3] Albuquerque E L, Loudon R and Tilley D R 1980 *J. Phys. C: Solid State Phys.* **13** 1775
[4] Mayer A P 1989 *J. Phys.: Condens. Matter* **1** 3957
[5] Marvin A, Bortolani V, Nizzoli F and Santoro G 1980 *J. Phys. C: Solid State Phys.* **13** 1607
[6] Bell J A, Zanoni R, Seaton C T, Stegeman G I, Bennett W R and Falco C M 1988 *Appl. Phys. Lett.* **52** 610
[7] Bottani C E, Ghislotti G and Mutti P 1994 *J. Phys.: Condens. Matter* **6** L85
[8] Ghislotti G and Bottani C E 1994 *Phys. Rev.* B at press
[9] Malischewsky P 1987 *Surface Waves and Discontinuities* (Amsterdam: Elsevier)
[10] Sagan H 1989 *Boundary and Eigenvalue Problems in Mathematical Physics* (New York: Dover) p 163
[11] Numerical Algorithms Group 1983 *NAG Fortran Library Manual* mark 10, v 2
[12] Bortolani V, Nizzoli F, Santoro G and Sandercock J R 1982 *Phys. Rev.* B **25** 3442
[13] Lekner J 1987 *Theory of Reflection* (Dordrecht: Martinus Nijhoff) p 5, 157
[14] Caporali R and Bottani C E 1994 to be published