Theory of Surface Acoustic Phonon Normal Modes and Light Scattering Cross Section in a Periodically Corrugated Surface

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We have developed a theory of surface acoustic phonon normal modes and of Brillouin-scattering ripple cross section for a periodically corrugated surface of an opaque material, in the small corrugation limit. Both the discrete and continuous spectra of acoustic modes on a grating have been studied within the elasticity theory. In the continuum, the Rayleigh wave becomes a resonance and hybridizes with the longitudinal pseudomode of the flat surface, giving rise to a gap. The theory explains quantitatively recent experimental results obtained for a shallow grating on a Si(001) surface.

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Recent advances in Brillouin light scattering (BLS) by acoustic surface modes in opaque materials include the study of periodically corrugated surfaces. Two problems have been investigated. The first one concerns BLS from metal gratings when surface plasmon polaritons are involved as intermediate states [1]; in this case the major theoretical interest [2] has been concentrated on the explanation of certain effective selection rules found in the experimental spectra taken with different polarizations of light. The second problem, which has been tackled experimentally only very recently [3], concerns the effects of periodicity on the surface phonon dispersion relations and BLS cross section. It is the latter problem we want to consider in this paper from the theoretical point of view.

There is a general physical interest behind this particular problem. First of all the presence of the corrugation implies the appearance of a surface Brillouin zone (BZ) and zone folding of the dispersion curves; this effect should not be mistaken with the well-known behavior of phonon branches in superlattices, where bulk modes are primarily observed [4]. Moreover, in the present case, there is a special interest in the folding of both a discrete spectrum of surface modes [Rayleigh wave (RW)] and a continuous spectrum of bulk modes projected on the surface and exhibiting some surface character, in particular the so-called pseudo surface modes [5].

In a very recent experimental paper [3] BLS has been actually used to investigate in detail the dispersion relation of surface and pseudo surface acoustic modes in a ion-milled holographic grating produced on the Si(001) surface. The periodicity of the grating was $a=2500$ Å and the depth $\zeta_0=175$ Å. The resulting corrugation strength $\zeta_0/a=0.07$ is considered to fall in the small roughness limit. Glass, Loudon, and Maradudin [6] explicitly showed that in such a limit, for a sinusoidal profile, the Rayleigh hypothesis and a perturbative expansion to first order in the corrugation strength yield good results for the surface wave dispersion curves. Indeed in the following we will rely on these approximations. The above mentioned paper [6], together with a subsequent one by these authors [7], offers a variety of calculations of surface and pseudo surface acoustic waves across gratings of different corrugation strengths in isotropic media, and it has been used by the experimentalists [3] as a guideline to interpret their Brillouin spectra qualitatively. However, a full understanding of BLS spectra needs not only the dispersion relations of the modes, but in addition the calculation of the normal modes and a proper theory for the light scattering cross section.

The purpose of this paper is to present a theoretical approach to the calculation of the surface acoustic normal modes of a continuous elastic medium and to the BLS cross section in a grating of an opaque material. We also apply the theory to the specific case of the Si(001) corrugated surface, which has been experimentally studied [3]. There are two possible approaches to the calculation of the normal modes on a grating: either to consider a complex surface wave vector $\mathbf{Q}$ so that surface and pseudo surface modes correspond to poles of the boundary conditions [7], or to keep $\mathbf{Q}$ real and to study in addition a continuous spectrum. We have chosen the second approach which naturally gives the normal modes and the surface power spectrum, to be compared directly with the experimental spectra.

Our system is made of a semi-infinite medium filling the region $z \le \zeta(x)$ and bound by a stress-free surface. The x axis is chosen along the phonon propagation direction; the z axis is normal to the flat surface. $\zeta(x)=\zeta_0 \times \cos(Gx)$ is the surface profile function; $G=2\pi/a$ defines the first surface BZ, with a being the grating periodicity and ζ_0 its depth. We assume a complete decoupling of the modes polarized in the x-z sagittal plane from the modes normal to it, as is the case for isotropic media and the (001) surface of cubic silicon with $\mathbf{Q}$ along the [110] x axis (case considered here and in the experiment [3]). The elastic displacement field in the medium is obtained in this geometry in terms of bulk modes as the Bloch solution

$$\mathbf{u}(x,z,t)=\mathbf{u}_i(x,z,t)+\mathbf{u}_d(x,z,t)\,, \tag{1}$$

where

$$\mathbf{u}_i(x,z,t)=\sum_\lambda A_\lambda^+(Q_i)\mathbf{e}_\lambda^+(Q_i)e^{iq_\lambda(Q_i)z}e^{iQ_ix}e^{-i\Omega t}$$

is the incident wave (of parallel wave vector Q_i) from $z\rightarrow-\infty$ to the surface (if any), and

$$\mathbf{u}_d(x,z,t)=\sum_{m,\lambda} A_\lambda^-(Q_m)\mathbf{e}_\lambda^-(Q_m)e^{-iq_{\lambda,m}z}e^{iQ_mx}e^{-i\Omega t} \tag{2}$$

is the reflected solution representing a traveling wave down to $z\rightarrow-\infty$ for $q_{\lambda,m}$ real, or an exponentially decaying wave if $q_{\lambda,m}=i\gamma_{\lambda,m}$ with $\gamma_{\lambda,m}>0$. In Eq. (2) $Q_m=Q_i+G_m$ and the sum over m extends to the reciprocal space $G_m=(2\pi/a)m$ determined by the surface profile $\zeta(x)$. The transverse momentum $q_{\lambda,m}\equiv q_\lambda(Q_m)$ is associated with the parallel momentum Q_m, such that the total momentum $(Q_m,\pm q_{\lambda,m})$ satisfies the Christoffel equation [5] for the bulk modes of frequency Ω whose polarization is specified by the vectors $\mathbf{e}_\lambda^\pm(Q_m)$. The index λ takes care of the two polarization modes in the x-z sagittal plane which we denote by $\lambda\rightarrow l,t$ resembling purely longitudinal and transverse solutions to which they reduce for an isotropic medium.

The bulk modes, i.e., the quantities $q_\lambda,\mathbf{e}_\lambda^\pm$, are calculated numerically while the unknowns $A_\lambda^\pm(Q_m)$ are determined imposing the stress-free boundary conditions at the surface $z=\zeta(x)$, i.e., $n_jT_{ij}=0$, with $i=1,3$. The surface normal is given by $\hat{\mathbf{n}}=(\hat{\mathbf{z}}-\zeta'\hat{\mathbf{x}})/(1+\zeta'^2)^{1/2}$, where $\zeta'=d\zeta/dx$ and $T_{ij}=c_{ijkl}\partial u_k/\partial x_l$ is the usual stress tensor. We use the Rayleigh hypothesis [6], i.e., we retain the expression in Eq. (1) valid up to the surface $z\rightarrow\zeta^-$. In the Fourier space one can write compactly the equivalent of the boundary conditions as

$$\sum_m \mathbf{H}^-(Q_n,Q_m)\mathbf{A}^-(Q_m)+\mathbf{H}^+(Q_n,Q_i)\mathbf{A}^+(Q_i)=0\,, \tag{3}$$

where $\mathbf{A}^\pm$ is the vector $(A^\pm_{\lambda=l},A^\pm_{\lambda=t})$ and $\mathbf{H}^\pm$ are 2×2 matrices defined as $\mathbf{H}^\pm=\mathbf{B}^\pm\mathbf{F}^\pm$, with $\mathbf{F}^\pm$ diagonal matrices,

$$F_{\lambda\lambda}^\pm(Q_n,Q_m)=a^{-1}\int_{-a/2}^{a/2}e^{\pm iq_\lambda(Q_m)\zeta(x)}e^{-i(Q_n-Q_m)x}dx$$

and

$$B_{1,\lambda}^\pm(Q_n,Q_m)=c_{55}\{\pm q_{\lambda,m}[\mathbf{e}_\lambda^\pm(Q_m)]_x+Q_m[\mathbf{e}_\lambda^\pm(Q_m)]_z\}+\frac{Q_m-Q_n}{q_{\lambda m}}\{\pm c_{11}Q_m[\mathbf{e}_\lambda^\pm(Q_m)]_x+c_{13}q_{\lambda,m}[\mathbf{e}_\lambda^\pm(Q_m)]_z\}\,,$$

$$B_{2,\lambda}^\pm(Q_n,Q_m)=c_{13}Q_m[\mathbf{e}_\lambda^\pm(Q_m)]_x\pm c_{33}q_{\lambda,m}[\mathbf{e}_\lambda^\pm(Q_m)]_z+\frac{Q_m-Q_n}{q_{\lambda,m}}c_{55}\{q_{\lambda,m}[\mathbf{e}_\lambda^\pm(Q_m)]_x\pm Q_m[\mathbf{e}_\lambda^\pm(Q_m)]_z\}\,.$$

When $\Omega<\Omega_t$ (discrete spectrum) there is no incident wave; therefore $\mathbf{A}^+=0$ and Eq. (3) allows one to study the dispersion relation of the surface mode in the discrete spectrum [6]. This method can be extended to the radiative region ($\Omega>\Omega_t$) neglecting $\mathbf{A}^+$ and solving the homogeneous system (3) using a complex $\mathbf{Q}$ or Ω; in this case the damping of the RW has been investigated [7]. Our purpose, however, is not limited to the calculation of the dispersion relations: We also need the eigenvectors and the line shape of the modes, in order to compare the theoretical spectra with the experimental data. In addition the method of Glass and Maradudin [7] presents numerical difficulties in the searching procedure of poles in the complex Ω plane, which probably are responsible for the lack of a gap opening, as will be shown later. Therefore in the radiative region we take fully into account the inhomogeneous term $\mathbf{A}^+$. When $\Omega_t<\Omega<\Omega_l$ Eq. (3) is solved for a single normal mode of the continuous spectrum. For $\Omega>\Omega_l$ and by taking for $\mathbf{A}^+$ the vectors

$$(0,1),(1,0)\,, \tag{4}$$

two normal modes are obtained. $\Omega_{t,l}=c_{t,l}Q_i$ are the transverse and longitudinal thresholds.

In the small corrugation limit the $\mathbf{H}^\pm$ matrices become

$$\mathbf{H}^\pm(Q,Q')\approx\delta_{Q,Q'}\mathbf{B}^\pm(Q,Q')+\zeta_{Q-Q'}\mathbf{M}^\pm(Q,Q')\,,$$

where $M_{j\lambda}^\pm(Q,Q')=\pm iq_\lambda(Q')B_{j\lambda}^\pm(Q,Q')$ and the Fourier transform of the sinusoidal grating profile is given by $\zeta_{Q-Q'}=(\zeta_0/2)\delta_{|Q-Q'|,G}$. In this limit Eq. (3), for $n=0$, becomes

$$\mathbf{A}^-(Q_i)=-(\mathbf{B}_i^-)^{-1}\{\mathbf{B}_i^+\mathbf{A}^+(Q_i)+\zeta_{Q_i-Q_R}\mathbf{M}^-(Q_i,Q_R)\mathbf{A}^-(Q_R)\}\,, \tag{5}$$

where $\mathbf{B}_i^\pm\equiv\mathbf{B}^\pm(Q_i,Q_i)$ and the wave vector $Q_R\equiv Q_{\pm1}$ is related to the RW folded from the nearest-neighbor Brillouin zones. In particular, in the range of energy we are dealing with, the only relevant term comes from Q_{-1}. It is precisely the presence in the latter equation of both bulk terms $\mathbf{A}^\pm(Q_i)$ and of the Rayleigh term $\mathbf{A}^-(Q_R)$ that makes possible the interaction between the surface longitudinal resonance (LR) of the flat surface and the folded RW. This is illustrated in the dispersion curves of Fig. 1 in the first surface BZ. The coefficients $\mathbf{A}^-(Q_R)$ appearing in Eq. (5) can be consistently calculated by solving the equation

$$\{\mathbf{B}_R^- -|\zeta_{Q_R-Q_i}|^2\mathbf{M}^-(Q_R,Q_i)(\mathbf{B}_i^-)^{-1}\mathbf{M}^-(Q_i,Q_R)\}\mathbf{A}^-(Q_R)+\zeta_{Q_R-Q_i}\{\mathbf{M}^+(Q_R,Q_i)-\mathbf{M}^-(Q_R,Q_i)(\mathbf{B}_i^-)^{-1}\mathbf{B}_i^+\}\mathbf{A}^+(Q_i)=0\,, \tag{6}$$

which has been derived from Eq. (3) with $n=-1$ in the small corrugation limit and using Eq. (5). The quantity $\mathbf{B}_R^- \equiv \mathbf{B}^-(Q_R,Q_R)$ is related to the surface mode for a flat surface which satisfies $\det(\mathbf{B}_R^-)=0$.

In Eq. (6) the grating shifts the Rayleigh pole to $\sim\zeta^2$, while Eq. (1) contains through Eq. (5) the characteristic Breit-Wigner behavior around the shifted pole. With the choice (4) the two solutions of Eq. (5) are not orthogonal, contrary to what happens in the flat-surface case. In order to get the cross-section formulas one has to orthogonalize them and normalize according to a standard procedure [8,9]. Equations (5),(6) form the starting point for the numerical calculations presented in this Letter. Retaining the ripple contribution [10] only, which is the relevant one in silicon [11], the spectral intensity is given by

$$\sigma(\Omega)\sim\frac{1}{\Omega^2}\sum_{Q',n}\delta(\Omega-\Omega_n)\left|a^{-1}\int_{-a/2}^{a/2}s^n(x;Q')e^{-i(Q-Q')x}dx\right|^2, \tag{7}$$

where Q is the exchange in the light parallel momentum $Q=K_i-K_f$ with $K_{i,f}=(\omega/c)\sin\theta_{i,f}$ and

$$s^n(x;Q')=\{W_z^n(x,z;Q')-\zeta'(x)W_x^n(x,z;Q')\}_{z=\zeta(x)}. \tag{8}$$

In the above equations we have used the nonmenclature of Ref. [8], n being the degeneracy and frequency index. Here $\mathbf{W}$ are orthonormal solutions obtained from Eq. (1). Equation (8) has been obtained projecting the acoustic field on the surface normal.

Figure 1 shows the experimental dispersion data of the acoustic modes for the Si(001) corrugated surface previously described, resulting from the peaks of the BLS spectra of Dutcher *et al.* [3]. The experimental points fall either in the discrete or in the continuous spectrum: The latter spectrum extends to frequencies above the dashed lines, representing the transverse threshold. The dotted lines mark the longitudinal threshold and also represent the dispersion curve of the LR of the flat surface previously mentioned. This is a pseudo or leaky mode which has been experimentally [12] and theoretically [13] studied with BLS for the first time in GaAs. The main features of the discrete spectrum are the gap opening at the zone boundary and the folding of the RW dispersion curve. In the continuous spectrum the experimental data exhibit the hybridization between the RW and the LR. The fact is remarkable because the LR of the flat surface is polarized parallel to the surface and therefore does not ripple the surface at all. Furthermore, in the previous calculations of Glass and Maradudin [7] no gap was foreseen.

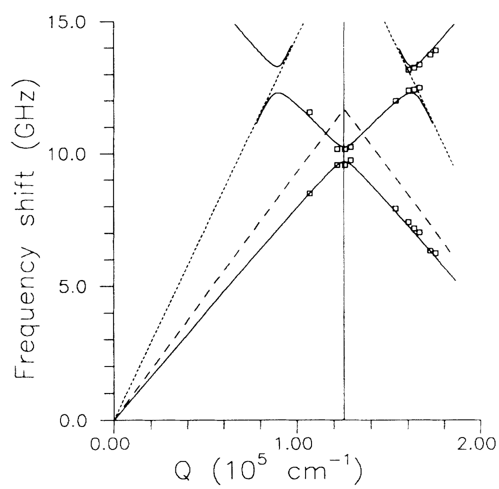

FIG. 1. Calculated dispersion relations for surface and pseudo surface modes for the Si(001) grating described in the text (solid lines). The experimental points are from Ref. [3] (squares). The slanting straight lines represent the transverse threshold (dashed lines) and the longitudinal threshold (dotted lines). The continuum of the radiative region extends above the dashed line. The vertical line ($Q=1.26\times10^5$ cm^{-1}) marks the surface zone boundary due to the grating.

Our theory and calculations explain quantitatively all these features. In Fig. 1 we compare the experimental and calculated dispersion relations of the surface modes and resonances. The agreement is excellent. The only input parameters are the grating data, already given at the beginning of the paper, and the elastic data of bulk silicon: $c_{11}=1.66$, $c_{12}=0.64$, $c_{44}=0.80$ 10^{11} N m^{-2}, $\rho=2330$ kg m^{-3}. The calculated dispersion curves in the continuous spectrum (above the dashed lines) have been drawn by taking the positions of the maxima in the cross section given by Eq. (7). In this part of the surface spectrum, not only the LR, but also the RW is a leaky or pseudomode. In particular, for Q in the second BZ, the coupling between RW and bulk modes is possible if $Q\geq G(1+c_R/c_t)^{-1}$, where c_R is the RW velocity. In this case the surface mode acquires a finite width $O(\zeta)$ and height $O(\zeta^{-1})$ and shifts to $\Omega_R\approx c_RQ+O(\zeta^2)$. For Q close to $G(1+c_R/c_l)^{-1}$ the coupling between the leaky RW and the LR gives rise to a gap, showing that the usual hybridization between normal modes of the same symmetry [14] occurs also in a continuum between pseudomodes. However, there is a peculiarity in this behavior: Resonances or pseudomodes have a natural linewidth $\Delta\Omega$, so that if they get closer than $\Delta\Omega$ they are no longer distinguishable and the gap practically vanishes. We have found that in the present case the gap disappears for a corrugation depth ζ_0 less than ~50 Å corresponding to a corrugation strength less than 2%.

We have now to explain why in proximity of the crossing between the RW and the LR the latter mode becomes observable in the spectra, although on the flat surface it is purely longitudinal. There are two reasons for that. First the surface boundary conditions mix the unfolded RW with the folded LR of the second zone so that the LR

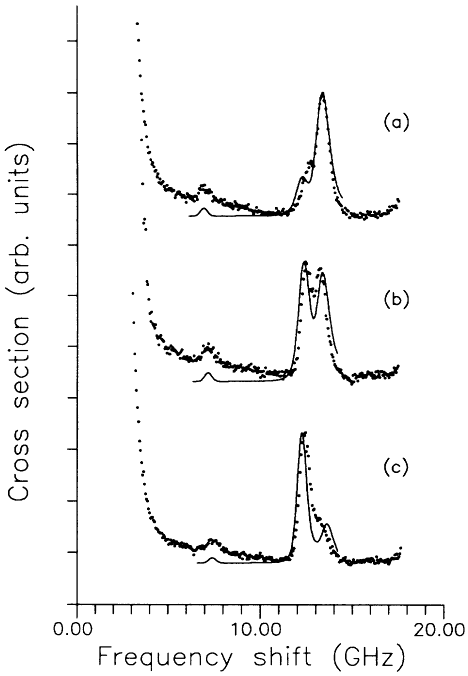

FIG. 2. Calculated (solid lines) and experimental [3] (dots) Brillouin spectra for the Si grating. The parallel wave vector Q is (a) 1.58, (b) 1.61, and (c) 1.64×10^5 cm^{-1}.

gains a z character: This contribution corresponds to the first term on the right-hand side of Eq. (8). Second, the ripple cross section given in Eqs. (7),(8) also contains a term proportional to $W_x^n \sim [\mathbf{e}_\lambda(Q_i)]_x A_\lambda^-(Q_i)$ which is resonant on the flat surface at the frequency of the LR for λ corresponding to the longitudinal partial wave. Actually we have found that both effects contribute to enhance the ripple cross section of the LR close to the hybridization. We conclude that the grating can rotate the polarization of the LR.

In Fig. 2 we show the comparison between the experimental and calculated BLS cross section for three interesting values of Q in the second BZ. Each spectrum shows three structures: At low frequency there is a small peak due to the folded RW (the unconvoluted cross section would exhibit here a δ function); at higher frequency there are two peaks belonging to the continuous spectrum. In order to compare directly with the experimental data, the theoretical curves have been convoluted with a Gaussian of half-width 0.74 GHz. The theory correctly describes the normal modes and the phonon-induced ripples close to the hybridization between the RW and the LR. In Figs. 2(a) and 2(c) the higher peak is due to the RW while the LR gives rise to a small shoulder; in Fig. 2(b) there are two equally high peaks corresponding to a complete hybridization between the RW and the LR. In conclusion we have shown that the surface acoustic phonon normal modes and the light scattering cross section in a shallow sinusoidal grating of an opaque material can be well described by a theory based on the Rayleigh hypothesis, perturbative expansion to first order in the corrugation strength, and the ripple scattering mechanism.

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