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Experimental evidence of the transformation of the Rayleigh surface phonon in $CaF_2/GaAs(1\bar{1}1)$ heterostructures of the accelerating type

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Abstract. The spectrum of surface acoustic phonons in films of CaF_2 on $GaAs(1\bar{1}1)$ has been investigated by means of Brillouin light scattering. We report the first experimental observation of the disappearance of the Rayleigh phonon with increasing film thickness in these structures of the *fast-on-slow* or *accelerating* type. For propagation along $\langle 121 \rangle$ at the disappearance of the Rayleigh surface phonon, a quasi-resonance in the continuum, whose spectral position tends to that of the Rayleigh phonon of $CaF_2(1\bar{1}1)$ for larger value of film thickness, is detected in the spectrum. Because of this behaviour we identify this quasi-resonance as the *pre-Rayleigh* mode of the film. When propagation is along $\langle 110 \rangle$ the role of the *pre-Rayleigh* mode of the film is played by the pseudosurface acoustic mode. The theoretical discussion is performed by comparing the Brillouin spectra with the surface projected phonon density of states for shear vertical phonons numerically calculated for $CaF_2/GaAs(1\bar{1}1)$ heterostructures.

1. Introduction

Brillouin scattering of light has been successfully used to study the spectrum of long-wavelength surface acoustic phonons in solid materials [1, 2]. Beside its fundamental significance, this type of spectroscopy represents a useful tool to investigate the dynamic elastic properties of solid surfaces as well as overlayers [3–5].

In anisotropic materials, for propagation along a generic crystallographic direction, the displacement field of surface acoustic phonons consists of a sagittal and a shear horizontal component. The sagittal plane is defined by the normal to the surface and the phonon propagation wavevector Q. The sagittal and shear horizontal motions are decoupled when propagation is along high-symmetry directions. In the rest of this paper we shall always refer to surface acoustic phonons that are polarized mainly in the sagittal plane.

The spectral analysis of surface acoustic phonons can be accomplished by the study of the surface projected phonon density of states SPPDS [6]. In the long-wavelength limit for a semi-infinite medium the spectrum of surface acoustic sagittal phonons is the union of a discrete and a continuous part. The latter begins at the *transverse threshold* $\Omega_t = QV_t$ where V_t is the quasi-shear vertical (SV) wave phase velocity of the material. The discrete spectrum is characterized by resonances in the SPPDS corresponding to true surface modes. In this frequency range, for a semi-infinite medium, only the Rayleigh wave (RW) exists.

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Above the transverse threshold quasi-resonances in the SPPDS can be found. These consist of a superposition of surface and bulk partial waves and may be thought of as surface acoustic waves that propagate with attenuation leaking energy into the bulk. Among these, the pseudosurface wave (PW) is of particular interest for this work [7]. Experimental evidence for the presence of the PW in the spectrum of GaAs($1\bar{1}1$) was given by Carlotti *et al* using Brillouin light scattering [8].

The presence of an overlayer modifies the spectrum of surface acoustic phonons for a semi-infinite medium. When the SV wave phase velocity of the layer V_{tl} is lower than that of the substrate V_{ts}, in the discrete spectrum further acoustic modes guided in the layer can exist (*Sezawa modes*). Instead, if $V_{tl} > V_{ts}$ the structure is of the *fast-on-slow* or *accelerating* type and in the discrete spectrum only the RW exists. In accelerating structures the RW phase velocity V_{RW} increases with the layer thickness h approaching the SV wave phase velocity of the substrate. When $V_{RW} > V_{ts}$ the RW is almost totally attenuated, being coupled to SV bulk waves and leaking its energy to the substrate.

Several experimental studies have been conducted on the spectrum of surface acoustic phonons in *slow-on-fast* structures [9, 10], while, to our knowledge, no experimental investigation in accelerating structures has yet appeared in the literature. In this work we have experimentally studied by means of Brillouin light scattering the spectrum of surface acoustic phonons of sagittal character in CaF_2/GaAs($1\bar{1}1$) heterostructures. These structures are of the accelerating type as shown in figure 1, where the phase velocity of the three bulk waves propagating parallel to a ($1\bar{1}1$) plane in CaF_2 and GaAs is plotted as a function of the propagation direction. In particular, we investigated the vanishing of the Rayleigh surface phonon with increasing layer thickness and how the disappearance is influenced by the presence of the pseudosurface phonon. Experimental spectra will be discussed with the aid of the SPPDS calculated numerically for the examined heterostructures using a Green function method.

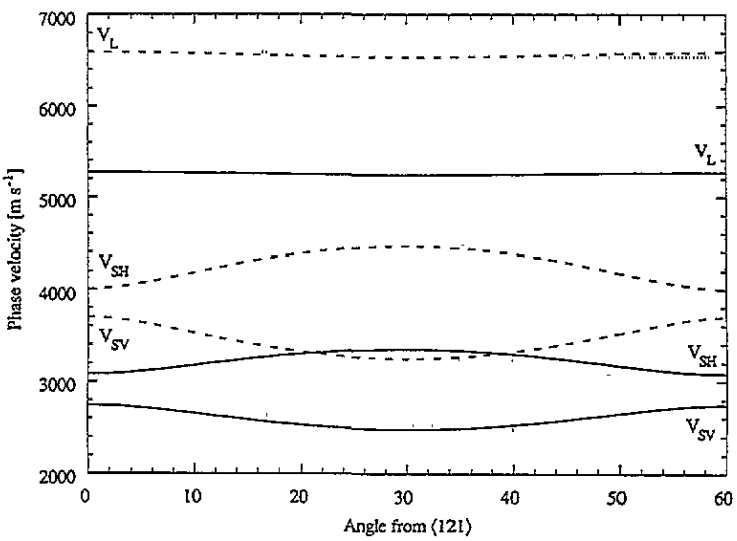

Figure 1. The phase velocity of the three bulk waves on a ($1\bar{1}1$) plane of CaF_2 (dashed line) and GaAs (full line: L, quasi-longitudinal wave; SH, quasi-shear horizontal wave; SV, quasi-shear vertical wave.

2. Experimental results

A series of CaF_2 films having thickness h in the range 10–550 nm was grown on GaAs($1\bar{1}1$) wafers by molecular beam epitaxy (MBE) [11]. The film quality was monitored *in situ* using reflection high-energy electron diffraction and a rare-earth ion photoluminescent probe [12].

Brillouin spectra were acquired in backscattering mode at room temperature using a 3+3 passes tandem interferometer having an overall finesse of approximately 100 [13]. Measurements were performed using incident p-polarized light from an argon ion laser oscillating in single longitudinal mode at $\lambda_0 = 514.5$ nm wavelength. No analysis of the polarization of the scattered field was conducted. The laser power incident on the sample surface was 150 mW and spectra were taken at a fixed angle of incidence ($\theta = 60°$). In backscattering the modulus of the transferred wavevector parallel to the surface is given by $Q = 2k_0 \sin\theta$ where $k_0 = 2\pi/\lambda_0$ is the modulus of the incident photon wavevector. To reduce the spectral broadening caused by the finite collection aperture a vertical slit was used to limit the collection angle $\Delta\theta$ to approximately 2°. This procedure has also the advantage of avoiding the splitting of peaks in the Brillouin spectrum caused by the interception of the backscattered photons by the mirror used to direct the incident light on the sample. With this configuration the measurement time, the same for all the spectra, was 5 h per spectrum.

Experimental Brillouin spectra of samples having increasing values of film thickness h for propagation along ⟨121⟩ and ⟨110⟩ are shown respectively in figure 2(*a*) and (*b*). The velocity units chosen for representing the horizontal axis are given by the relationship $V = \Omega/Q$ where Ω is the experimental frequency shift. When propagation is along ⟨121⟩ for a ($1\bar{1}1$) surface of a cubic crystal the shear horizontal and sagittal motions are decoupled. The RW polarized in the sagittal plane is the sole surface acoustic mode present in the spectrum when the film thickness h is in the range 0–50 nm. With increasing film thickness the RW phase velocity tends to the substrate SV wave phase velocity (the vertical dashed line in figure 2(*a*)) until it reaches it at around $h = 120$ nm. At this value of h the RW peak vanishes and only a very broad peak in the continuum is visible in the spectrum. Above $h = 120$ nm this evolves as a sharp peak in the continuum and shifts its spectral position tending to the RW phase velocity of the $CaF_2(1\bar{1}1)$ film.

When propagation is along ⟨110⟩ the shear horizontal and sagittal motions are coupled and both the RW and PW are present in the spectrum of GaAs($1\bar{1}1$) (figure 2(*b*), $h = 0$ nm). These two peaks are observable also for film thickness up to $h = 50$ nm. At this value of h the RW phase velocity is higher than the substrate SV wave phase velocity and the RW peak decreases in intensity and widens. At $h = 120$ nm the RW peak completely disappears while the PW is broader but still observable. Above $h = 120$ nm the PW peak sharpens and shifts toward the RW velocity of $CaF_2(1\bar{1}1)$.

The low intensity of the RW and PW peaks observable in the spectra at $h = 20$ nm is probably due to interference effects in the Brillouin scattering cross section. As a matter of fact, both CaF_2 and GaAs are partly transparent to the optical radiation used in the experiments and interference effects between the ripple and elasto-optic contributions to the cross section might be expected [10].

3. Discussion

A theoretical discussion of the character of surface acoustic sagittal phonons in accelerating structures is performed by means of the long-wavelength limit of the SPPDS. This quantity

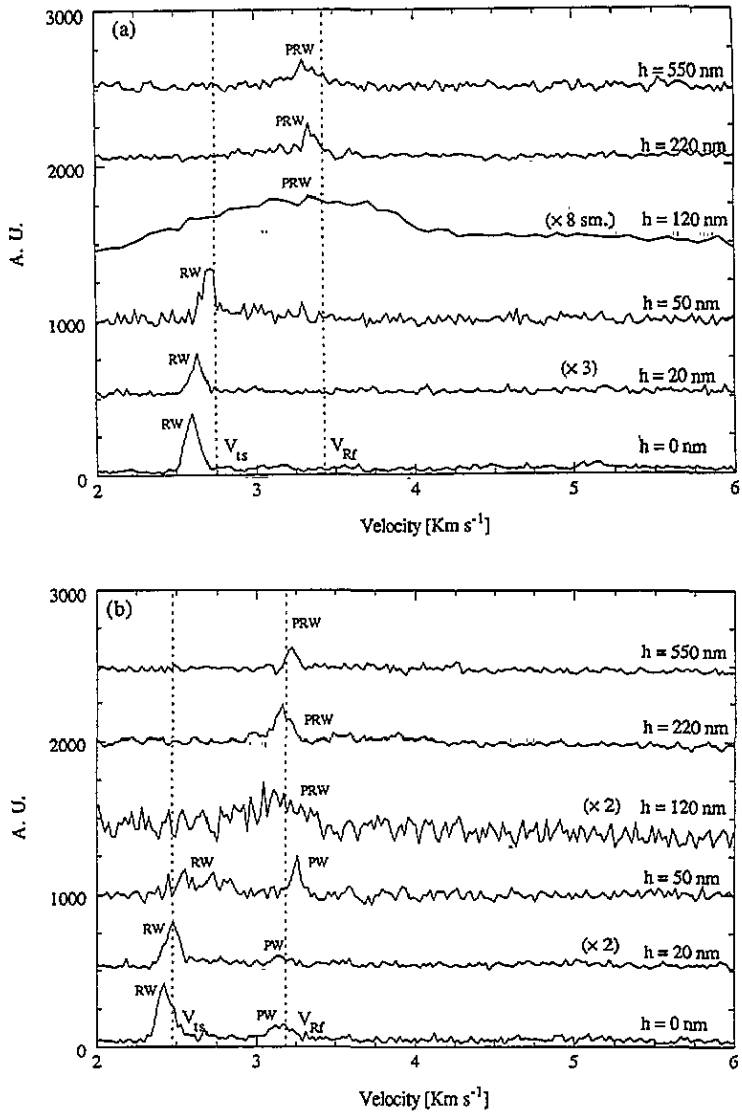

Figure 2. Experimental Brillouin spectra of $CaF_2/GaAs(1\bar{1}1)$ heterostructures with varying values of film thickness h: (*a*) propagation along ⟨121⟩; (*b*) propagation along ⟨110⟩. The SV wave phase velocity (V_{ts}) in the GaAs substrate and the Rayleigh wave phase velocity (V_{Rf}) in the CaF_2 film are shown as vertical dashed ines; RW, Rayleigh wave; PW, pseudosurface wave; PRW pre-Rayleigh wave (sm., smoothed curve).

is proportional to the power spectrum of the displacement component normal to the surface evaluated at the free surface. The SPPDS has been numerically calculated for $CaF_2/GaAs(1\bar{1}1)$ heterostructures using a Green function method, which is briefly outlined in the following [14, 15].

The equations of motions in the bulk of the substrate and overlayer for the dynamical Green tensor $D_{ij}(x, x'; \omega)$ are

$$\left(\sum_j \delta_{ij}\omega^2 + \frac{1}{\rho}C_{ijkl}\frac{\partial^2}{\partial x_j \partial x_l}\right) D_{ik}(\boldsymbol{x}, \boldsymbol{x}'; \omega) = \delta_{ik}\delta(\boldsymbol{x} - \boldsymbol{x}') \qquad (i, j, k, l = 1, 2, 3) \qquad (1)$$

where C_{ijkl} and ρ are within each material the elastic constant tensor and the mass density. Because the system is translationally invariant in directions parallel to the surface, it is convenient to Fourier analyse the dynamical Green tensor $D_{ij}(\boldsymbol{x}, \boldsymbol{x}'; \omega)$ and to deal with the Fourier component $D_{ij}(x_3, x_3'; \omega, Q)$. Taking the x_3 axis normal to the surface pointing toward the substrate, the boundary conditions become

$$[D_{ij}(x_3, x_3'; \omega, Q)]_{x_3=0^+} = [D'_{ij}(x_3, x_3'; \omega, Q)]_{x_3=0^-} \qquad (2a)$$

$$\left[\sum_k B_{ik}(x_3; Q) D_{kj}(x_3, x_3'; \omega, Q)\right]_{x_3=0^+} = \left[\sum_k B'_{ik}(x_3; Q) D'_{kj}(x_3, x_3'; \omega, Q)\right]_{x_3=0^-} \qquad (2b)$$

(continuity of displacements and normal stresses at the interface placed at $x_3 = 0$),

$$\left[\sum_k B_{ik}(x_3; Q) D_{kj}(x_3, x_3'; \omega, Q)\right]_{x_3=-h} = 0 \qquad (3)$$

(stress-free boundary conditions at the free surface placed at $x_3 = -h$) where

$$B_{ik}(x_3; Q) = \frac{1}{\rho}\sum_l C_{i3kl}\left((1 - \delta_{l3})\mathrm{i}Q_l + \delta_{l3}\frac{\mathrm{d}}{\mathrm{d}x_3}\right) \qquad (4)$$

and $Q = Q_1\boldsymbol{x}_1 + Q_2\boldsymbol{x}_2$ ($\boldsymbol{x}_1$ and $\boldsymbol{x}_2$ are the unit vectors in the 1 and 2 directions). Primed (unprimed) quantities refer to the layer (substrate). $D'(D)$ stands for the Green function when the point x_3 is in the layer (substrate). The calculations were performed taking the x_1 axis parallel to the propagation wavevector Q and using the rotation matrix indicated in [14] and [15] to transform the $D_{ij}(x_3, x_3'; \omega, Q)$.

The *smoothed* surface projected phonon density of states $n_{ij}(\omega; Q)$ has been evaluated by the surface projection of the Green function $D_{ij}(x_3 = 0,\ x_3' = 0; \omega, Q)$ using the relationship

$$n_{ij}(\omega; Q) = (1/2\pi\mathrm{i})[D_{ij}(x_3 = 0, x_3' = 0; \omega - \mathrm{i}\varepsilon, Q) - D_{ij}(x_3 = 0, x_3' = 0; \omega + \mathrm{i}\varepsilon, Q)]. \qquad (5)$$

Equations (1) have been numerically solved for propagation along ⟨121⟩ aned ⟨110⟩ in a film of CaF_2 on GaAs($1\bar{1}1$). Calculations have been performed at the same values of film thickness as used in the experiments. According to a standard procedure, in the computation of equation (5) a small imaginary part ε in frequency has been introduced.

To discuss the sagittal part of the SPPDS we considered only the $n_{33}(\omega; Q)$ because, for the propagation directions considered in this work, the $n_{11}(\omega; Q)$ exhibits the same frequency behaviour. Results of the calculations of the $n_{33}(\omega; Q)$ for propagation along ⟨121⟩ and ⟨110⟩ are shown respectively in figure 3(*a*) and (*b*). When propagation is along ⟨121⟩ at the disappearance of the RW peak (figure 3(*a*), $h = 120$ nm) a very broad quasi-resonance in the continuum is visible. With increasing film thickness, the intensity of this quasi-resonance gradually rises, becoming a very sharp maximum when $h = 550$ nm. This sharp quasi-resonance in the continuum is detected as a peak in the experimental spectrum (figure 2(*a*), $h = 550$ nm). The spectral position of the quasi-resonance approaches the RW

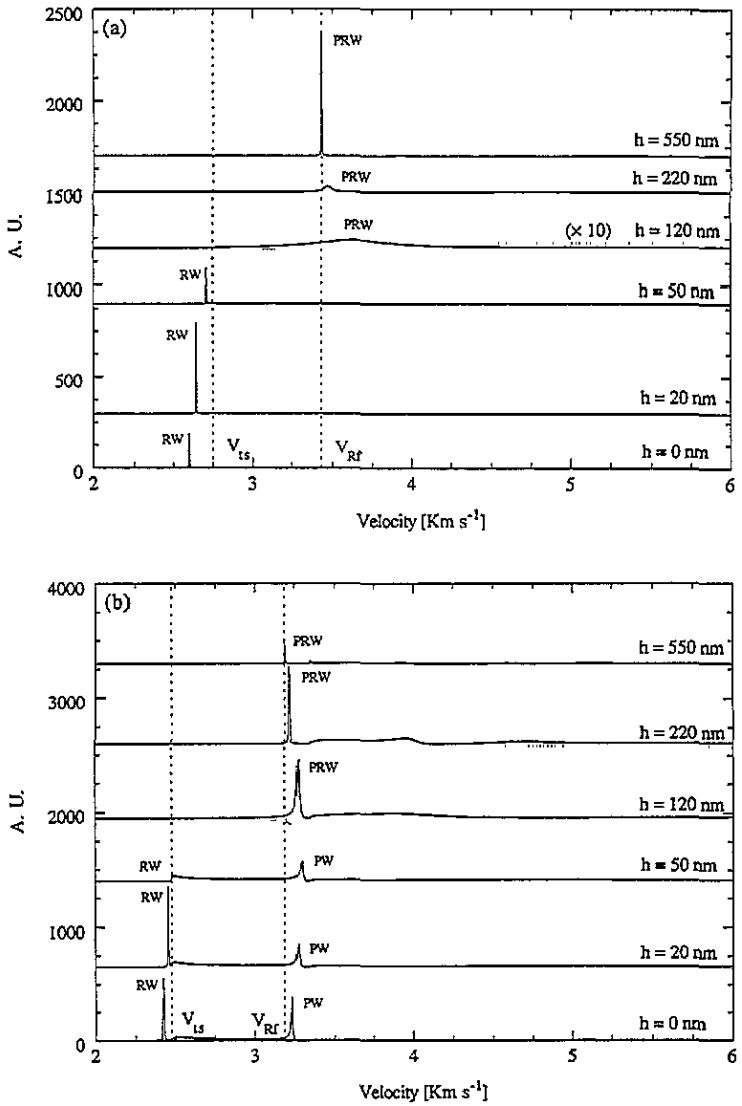

Figure 3. The normal component $n_{33}(\omega; Q)$ of the surface projected density of states for $CaF_2/GaAs(1\bar{1}1)$ heterostructures with varying values of film thickness h: (*a*) propagation wavevector parallel to ⟨121⟩; (*b*) propagation wavevector parallel to ⟨110⟩. Calculations were performed taking the small imaginary part ε in frequency equal to $2\pi 13$ MHz. The labels and vertical dashed lines have the same meaning as in figure 2.

phase velocity of $CaF_2(1\bar{1}1)$ with increasing film thickness. Along ⟨121⟩ the quasi-resonance experimentally observed in the continuum is therefore interpretable as the *pre-Rayleigh* mode of the film.

For propagation along ⟨110⟩ the situation is different (figure 3(*b*)). At the disappearance of the RW resonance, the PW, present in the SPPDS also for vanishing values of h, still exists. At higher h values, the PW quasi-resonance increases in intensity and shifts its position toward the RW phase velocity of $CaF_2(1\bar{1}1)$. Therefore along ⟨110⟩ the PW plays the role of the *pre-Rayleigh* mode of the film. The small peak observable at around 3.3–3.4 km s^{-1}

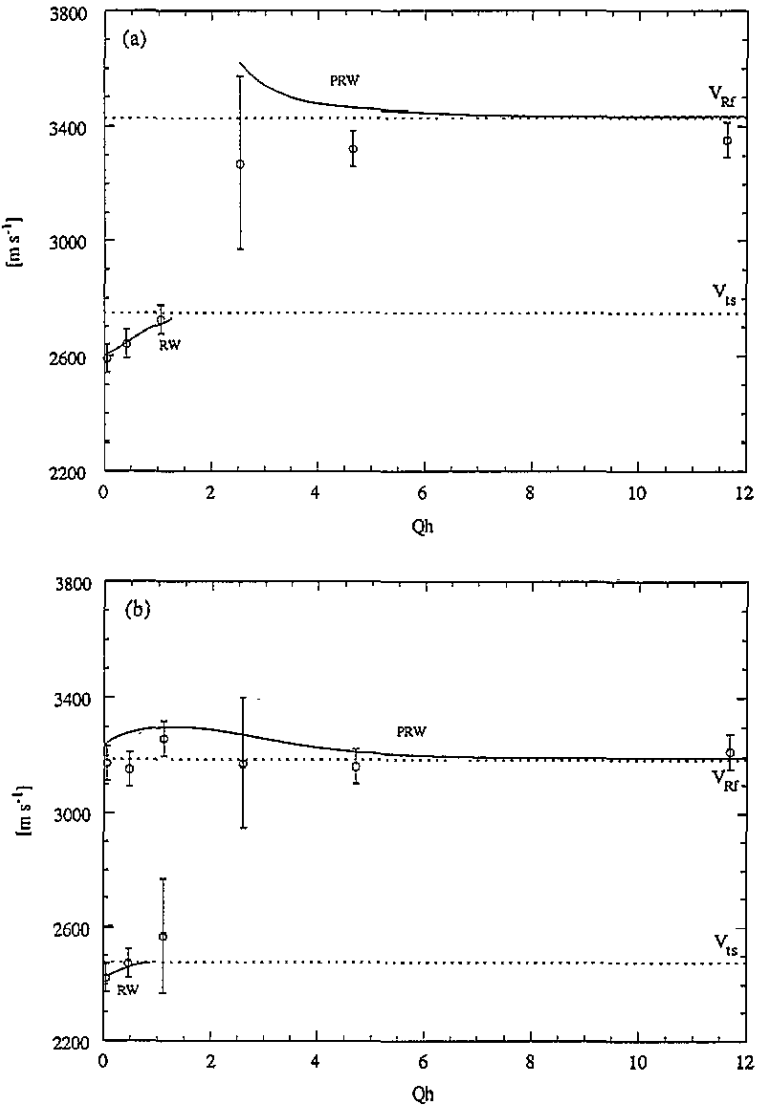

Figure 4. Experimental phase velocity of the Rayleigh wave (RW) and pre-Rayleigh wave (PRW) *of the* CaF_2 *film as a function of the* Qh *product compared with the dispersion curves calculated* from the SPPDS: (*a*) propagation along ⟨121⟩; (*b*) propagation along ⟨110⟩. The SV wave phase velocity (V_{ts}) in the GaAs substrate and the Rayleigh wave phase velocity (V_{Rf}) in the CaF_2 film are shown as dashed lines. Error bars were obtained by the FWHM of the Brillouin peaks.

in the SPPDS calculated at the larger film thickness ($h = 550$ nm) could be considered as the *pre-pseudosurface* mode of the film. This peak is not detected in the corresponding experimental spectrum.

To summarize our results we compared the experimental RW and PW phase velocity plotted as a function of the Qh product with the theoretical curves obtained by the spectral positions of the RW resonance and PW quasi-resonance in the SPPDS (figure 4). A reasonable agreement of experiments with theory is found for modes in the discrete part of the spectrum. In the continuum the experimental phase velocity values, although reproducing the behaviour

of the theoretical results, are lower mainly for propagation along ⟨121⟩.

4. Conclusions

By means of Brillouin light scattering we studied the long-wavelength spectrum of surface acoustic phonons in accelerating structures. The examined materials consisted of $CaF_2/GaAs(1\bar{1}1)$ heterostructures grown by MBE. In these structures we report the first experimental observation of the disappearance of the Rayleigh surface phonon when its phase velocity approaches that of the quasi-shear vertical wave of the substrate. For propagation along ⟨121⟩, in correspondence with the disappearance of the Rayleigh surface phonon, a peak in the continuum is detected in the spectrum. This peak corresponds to a broad quasi-resonance in the SPPDS whose spectral position tends to the Rayleigh surface phonon of $CaF_2(1\bar{1}1)$ for larger values of film thickness. Because of this behaviour we identified this quasi-resonance as the pre-Rayleigh mode of the film. When propagation is along ⟨110⟩, the PW plays the role of the pre-Rayleigh mode of the film. These conclusions were drawn by comparing the experimental spectra with the SPPDS of SV phonons calculated at the same values of film thickness used in the experiments.

Acknowledgments

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