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Localized surface phonon propagation in CaF_2/Si(110) heterostructures

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Abstract. We have studied experimentally the propagation of the surface phonon modes in CaF_2/Si(110) heterostructures with $h = 0$, 10 and 200 nm thickness by Brillouin light scattering spectroscopy. The angular azimuthal behaviour of the Rayleigh mode was found to be close to that of the material of the substrate in the case with $h = 10$ nm, and to that of the layer in the case with $h = 200$ nm. In an $h = 200$ nm heterostructure for the azimuthal directions close to $[1\bar{1}0]$ it was concluded that the behaviour of the first localized mode is described well by the proper pseudo-surface mode of CaF_2 of the (110) plane, whereas higher-mode (i.e. second, third) velocity values were found to be close to that of the material of the substrate. Experimental and calculated velocity values match each other rather well.

1. Introduction

Low-dimensional objects are acquiring ever-growing significance in the physics of solid state. Noteworthy among these are the surfaces of crystalline or amorphous specimens and layered structures grown on top of them. Many physical properties of such structures are associated with the behaviour of the surface phonons.

Brillouin light scattering spectroscopy is known to be an effective tool for long-wavelength surface phonon spectrum analysis [1, 2]. In addition to the probing of the Rayleigh mode (RM) phonon component it enables one to study some surface phonon peculiarities of bulk origin, i.e. leaky or pseudo-surface phonon modes characterized by their wave vector being inclined from the surface into the medium [3], see [1, 2, 4–9].

The presence of a film on the solid surface, provided that certain relations between the elastic properties of the layer/substrate materials hold, modifies the initial surface phonon distribution, manifesting itself in RM velocity reduction as well as in the appearance of higher-order phonon modes, of Love and Sezawa type, localized near the surface [2, 10–13].

Brillouin studies of layered structures are mainly focused on objects based on isotropic materials, or on the trivial cases of simple azimuthal direction propagation in high-symmetry planes.

However, the elastic anisotropy of the constituents should influence the RM and local mode propagation behaviour. This effect can be especially pronounced when the divergence of the elastic properties of the materials involved results in qualitatively different sets of acoustical phonon modes. The CaF_2/Si(110) heterostructure may be considered as an object of this sort.

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Both of the constituents are of cubic symmetry, their respective elastic anisotropy parameters $\mu = 2C_{44}/C_{11}-C_{12})$ being equal to 1.57 (Si) and 0.53 (CaF_2) [3, 14]. Here C_{ij} are the elastic moduli.

The elastic properties of bulk Si (substrate material) and CaF_2 (layer material) in the (110) plane are described by the curves of figure 1.

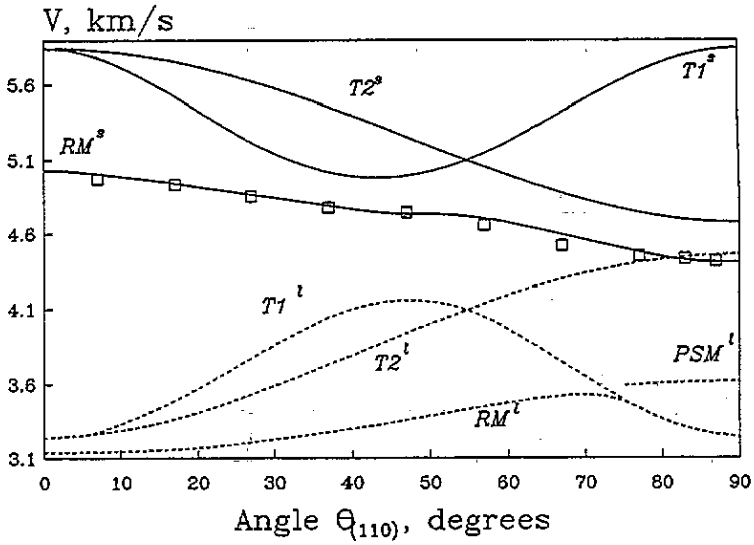

Figure 1. Si and CaF_2 angular dependence of surface and bulk (transverse) acoustic velocity values on $\theta_{(110)}$ in the (110) plane. Here $\theta_{(110)}$ is the angle between the $[00\bar{1}]$ crystallographic direction and the orientation of the sagittal plane. Si: full RM^s, $T1^s$, $T2^s$ lines, theoretical curves; squares, Brillouin spectroscopy measured velocity values. CaF_2: dashed RM^l, $T1^l$, $T2^l$, PSM^l lines, theoretical curves. The curves were calculated using the elastic moduli of [14].

In the case of Si the Rayleigh mode (RM^s) shows a monotonic velocity decrease from 5.025 km s^{-1} to 4.408 km s^{-1} as $\theta_{(110)}$ varies from 0° (RM^s wave vector q_{RM} parallel to the $[00\bar{1}]$ crystallographic direction) to 90° ($q_{RM} \parallel [1\bar{1}0]$). The plane of the RM^s displacement ellipse is orthogonal to the (110) surface for all $\theta_{(110)}$ values and coincides with the sagittal plane at $\theta_{(110)} = 0°, 90°$ [3]. The angular dependence of the velocities of both bulk shear waves ($T1^s$ and $T2^s$) in the (110) plane is also depicted in figure 1.

Corresponding theoretical curves RM^l, $T1^l$, and $T2^l$ for the (110) plane of the CaF_2 crystal are shown in figure 1 by dashed lines. In this case the RM^l displacement ellipse lies in the sagittal plane only at $\theta_{(110)} = 0° (q_{RM} \parallel [00\bar{1}])$. As one leaves the $[00\bar{1}]$ propagation direction, the RM^l displacement ellipse plane becomes non-parallel to the sagittal plane. Finally, for the azimuth directions close to the $[1\bar{1}0]$ ($\theta_{(110)} = 90°$) RM^l gradually degenerates into a linearly in-plane polarized shear wave. In addition, a certain region of azimuthal directions close to $[1\bar{1}0]$ is characterized by the presence of a pseudo-surface wave branch (PSM^l, see $\theta_{(110)} = 75$–$90°$). The latter is found to be of 'pure' Rayleigh wave character at $\theta_{(110)} = 90°$ [3].

Obviously, surface acoustical properties of the substrate and film differ dramatically. For the substrate surface one predicts monotonic dependence of wave velocity on azimuth, and at the same time the presence of the pseudo-surface wave in the vicinity of $[1\bar{1}0]$ for the (110) surface of the film material is expected. These 'contradictory' properties of the constituents should affect propagation behaviour of the Rayleigh wave and higher-order modes in the CaF_2/Si(110) system.

In order to describe the mechanical properties of the CaF_2Si(110) heterostructure, the propagation of the surface phonon system as a function of (i) the azimuthal direction of their propagation, and (ii) film thickness, h, should be examined.

CaF_2/Si(110) specimens used in our experiments were grown by molecular beam epitaxy in a research chamber [15]. The perfection of the film lattice structure as well as its surface morphology was properly monitored using the high-energy electron diffraction (HEED) method [15].

Brillouin light scattering spectra associated with surface phonon excitations have been registered by a Burleigh system including a five-pass piezoscanned Fabry–Perot interferometer of typical finesse $\sim$ 60 and of a contrast $\geqslant 10^{10}$, and a Spectra-Physics 165-03 single-frequency argon ion laser (l = 514.5 nm line). The power of the incident light beam was limited to 50 mW in order to protect samples from damage. Measurements were taken at room temperature in backscattering geometry with both the incident and detected light beams polarized in the plane of incidence (p–p scattering; for details see also [16]).

Two theoretical approaches were used for surface phonon component description. One method employs algorithms [17] based on the surface Green function matching method [4, 18]. The other uses Farnell's equations of motion routines [3, 10]. The velocity values obtained by these two procedures are always the same.

2. Results and discussion

Brillouin satellite frequency shifts δf versus azimuthal direction in the (110) free surface of Si are shown in figure 1 as squares. Frequency shifts were properly recalculated into velocity units according to the relation $V = \delta f l/(2\sin\alpha)$, where α is the angle of light incidence.

Experimental velocity values coincide with theoretical ones.

CaF_2 is characterized by a strong elasto-optic coupling that masks the light scattered from the surface phonons in the case of the bulk samples. At the same time, measurements conducted by us with opaque PbS and PbTe crystals with similar elastic anisotropy parameter values to CaF_2, $\mu = 0.307$ (PbTe), 0.508 (PbS) [3, 14] have shown that for the directions close to $[00\bar{1}]$, the principal Brillouin satellite frequency shift corresponds to the RM, whereas for the $[1\bar{1}0]$ direction the Brillouin line position corresponds to the pseudo-surface mode branch. This is due to the specific sensitivity of Brillouin surface p–p light scattering mainly to the normal-oriented surface excitations as well as by characteristic azimuthal displacement behaviour of both the Rayleigh and the leaky modes [2, 19, 20].

CaF_2/Si(110) heterostructure azimuthal dependences of elastic excitation velocities are shown in figure 2 (h = 10 nm) and figure 3 (h = 200 nm). Points (squares) indicate the Brillouin satellite positions, and continuous lines correspond to the calculated velocity values. Dotted lines here are similar to RM^s, $T1^s$, $T2^s$, RM^l, $T1^l$, and $T2^l$ of figure 1.

In the case of the system with h = 10 nm, see figure 2, the presence of a film leads to a minor (approximately 2.5%) proportional reduction of the RM velocity for all $\theta_{(110)}$ values relative to the free Si(110) plane sample surface, the RM displacement behaviour being perturbed insufficiently. As one can see, the experimental data fit the corresponding theoretical curve quite well.

For the samples with h = 200 nm layer thickness, the surface phonon distribution is found to be strongly modified by the presence of the film (see figure 3). Here film-localized

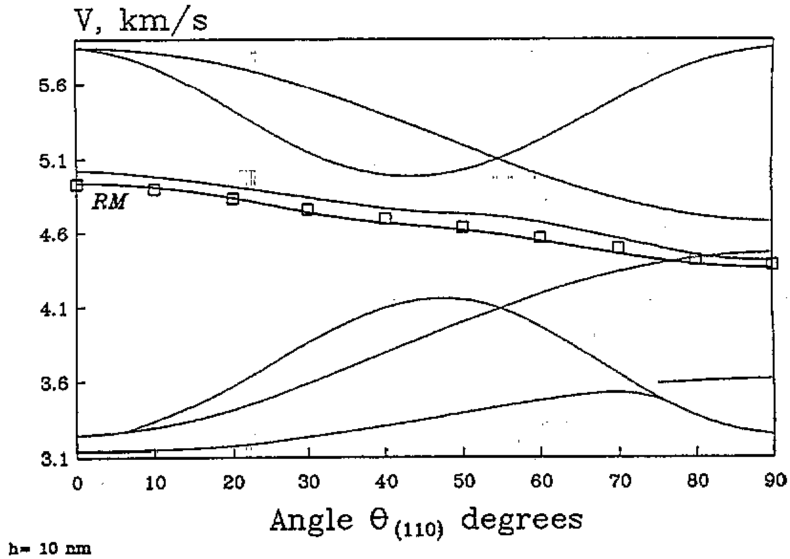

Figure 2. RM velocity values for different azimuthal propagation directions in the CaF_2/Si(110) heterostructure with h = 10 nm: full line, RM theoretical curve; squares, measured velocity values. The RM^s, $T1^s$, $T2^s$, RM^l, $T1^l$, $T2^l$, and PSM^l theoretical curves of figure 1 are depicted here by dotted lines. The curves were calculated using the elastic moduli of [14].

surface phonon modes appear (see curves 1, 2 and 3), and the initial RM velocity decreases so it may now be estimated well by the respective RM^l values in the whole $\theta_{(110)}$ region. That is, calculations show that at $\theta_{(110)} = 0°$ RM particle displacement lies in the sagittal plane as expected for the (110) free surface of CaF_2. With $\theta_{(110)}$ variation from 0° to 90° the RM displacements gradually change their orientation from sagittal to parallel to the (110) plane as occurs in the material of the layer.

The first localized phonon mode also belongs to the specific CaF_2 velocity region (see mode 1, figure 3). In the 80–90° range of $\theta_{(110)}$ this mode, being initially (at $\theta_{(110)} = 0°$) of the Love mode type, changes its displacement orientation to the sagittal one. One should note that for these $\theta_{(110)}$ values mode 1 eventually substitutes the pseudo-surface mode of the substrate material.

This specific azimuthal behaviour of the displacements of the RM and the lower localized mode substantially influences the spectral content of surface p–p scattered light. A good correlation may be seen between low-frequency Brillouin satellite positions and the RM branch in $\theta_{(110)} = 0°$ environs, where the RM induces noticeable normal surface deformations. No Brillouin peaks corresponding to the RM were detected at $\theta_{(110)} = 90°$, p–p scattering being practically insensitive to shear-oriented surface excitations. On the contrary, in the case of the first mode Brillouin peak positions correlate well with those calculated only for azimuthal directions close to [110] ($\theta_{(110)} = 90°$) where its surface displacements have reasonable normal projections. The authors of the present publication are not aware of analogous observations of film proper mode behaviour that can be described asymptotically by a pseudo-surface acoustical mode of the film material.

The higher modes of the structure (see modes labelled 2 and 3) are relatively more mixed in terms of their displacement character for all intermediate $\theta_{(110)}$ values, being purely Sezawa and Love modes at $\theta_{(110)} = 0°$ (for 3 and 2, respectively), and at $\theta_{(110)} = 90°$ (for 2

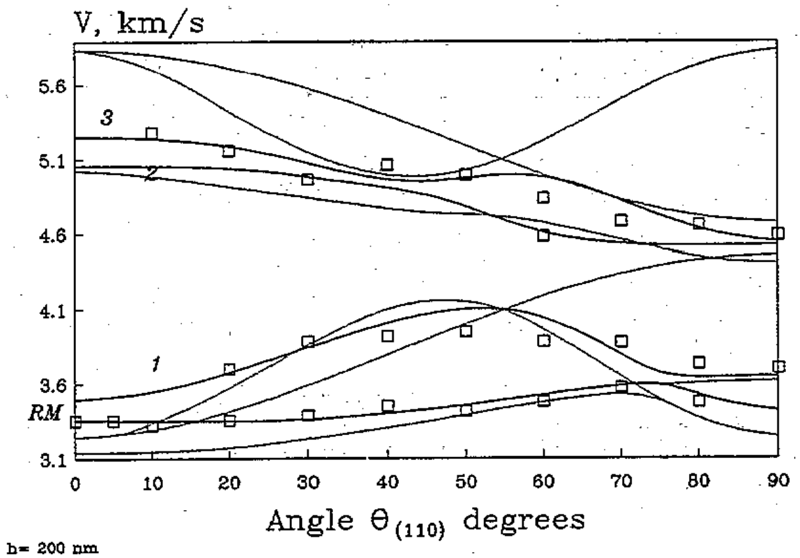

Figure 3. RM and localized mode velocity values for different propagation directions in the CaF_2/Si(110) heterostructure with $h = 200$ nm: full line, RM and mode 1, 2 and 3 theoretical curves; squares, measured velocity values. The RM^s, $T1^s$, $T2^s$, RM^l, $T1^l$, $T2^l$, and PSM^l theoretical curves of figure 1 are depicted here by dotted lines. The curves were calculated using the elastic moduli of [14].

and 3, respectively). The latter results in the detection of only the third mode at azimuthal directions close to $\theta_{(110)} = 0°$.

It should be noted that while the angular dependence of the velocity of the lower mode tends to that of the material of the film, the higher ones behave in general similarly to the surface waves of the substrate. The latter behaviour is due to their weaker degree of localization near the surface compared with the RM and, consequently, greater influence of the properties of the substrate on the higher-mode propagation characteristics. In this sense the higher modes should be more sensitive to the elastic properties and the structural perfection of the interface region.

The discrepancies between calculated velocity values and those obtained experimentally are found to be nearly the same for surface acoustic waves of different orders. This proves the homogeneity of the elastic properties of the specimens studied along their thickness, and the absence of substantial structural distortions in the interface region.

In all the experiments Brillouin satellite linewidths were properly controlled using the spectral deconvolution procedures of [21]. Experimental accuracy being insufficient, no broadening of the Brillouin peaks was detected. The latter did not permit us to measure the attenuation of the surface phonon modes.

3. Conclusion

Long-wavelength surface phonon excitation spectra of CaF_2/Si(110) heterostructures with $h = 0$, 10 and 200 nm film thickness have been studied by means of Brillouin light scattering spectroscopy. Azimuthal dependences of the surface mode velocities in the (110) plane have been examined.

In the case of $h = 10$ nm the azimuthal dependence of the RM closely resembles that of the Si(110) free sample surface with $\sim 2.5\%$ reduction of the velocity value. For a film with $h = 200$ nm the azimuthal dependence of Rayleigh wave velocity and that of the first localized mode tend to the corresponding dependences of the film material. For the azimuthal directions close to [110] the first mode behaves similarly to the pseudo-surface mode of the substrate material.

The comparison of experimental and theoretical values of the surface mode velocity allows one to conclude on the long-range homogeneity of the specimens inspected.

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References

[1] Sandercock J R 1976 *Solid State Commun.* **26** 547–51
[2] Nizzoli F and Sandercock J R 1990 *Dynamical Properties of Solids* vol 6, ed H K Horton and A A Maradudin (Amsterdam: North-Holland) pp 281–335
[3] Farnell G W 1970 *Physical Acoustics* vol VI, ed W P Mason and R N Thurston (New York: Academic) pp 109–66
[4] Velasco V R and Garcia-Moliner F 1980 *J. Phys. C: Solid State Phys.* **13** 2237–56
[5] Aleksandrov V V, Velichkina T S, Mozhaev V G and Yakovlev I A 1991 *Solid State Commun.* **77** 559–63
[6] Carlotti G, Fioretto D, Giovanni D, Nizzoli F, Socino G and Verdini L 1992 *J. Phys.: Condens. Matter* **4** 257–62
[7] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Zh. Eksp. Teor. Fiz.* **102** 1891–7 (Engl. transl. 1992 *Sov. Phys.–JETP* **102** 1018–21)
[8] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Phys. Lett.* **171A** 103–6
[9] Aleksandrov V V, Vorob'ev P A, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Preprint* 9/1992 Physics Department, Moscow State University, pp 1–19
[10] Farnell G W and Adler E L 1972 *Physical Acoustics* vol IX, eds W P Mason and R N Thurston (New York: Academic) pp 35–127
[11] Zanoni R, Bell J A, Stegeman G I and Seaton C T 1987 *Thin Solid Films* **154** 225–37
[12] Hillebrands B, Lee S, Stegeman G I, Cheng H, Pots J E and Nizzoli F 1989 *Surf. Sci.* **211/212** 387–93
[13] Karanikas J M, Sooryakumar R and Phillips J M 1989 *J. Appl. Phys.* **65** 3407–10
[14] *Landolt-Börnstein New Series* 1966 Group III, vol 1 (Berlin: Springer) p 1983
[15] Gastev S V, Novikov S V, Sokolov N S and Yakovlev N L 1987 *Pis. Zh. Tekh. Fiz.* **13** 961–4 (Engl. transl. 1987 *Sov. Tech. Phys. Lett.* **13** 401–3)
[16] Aleksandrov V V, Velichkina T S, Voronkova V I *et al* 1989 *Phys. Lett.* **142A** 307–11
[17] Vorob'ev P A and Aleksandrov V V private communication
[18] Sanz-Velasco E, Hardin-Duparc O and Velasco V R 1983 *Surf. Sci.* **126** 202–7
[19] Aleksandrov V V, Velichkina T S, Potapova Ju B and Yakovlev I A 1992 *Phys. Lett.* **170A** 165–70
[20] Aleksandrov V V, Vorob'ev P A, Velichkina T S, Potapova Ju B and Yakovlev I A 1993 *Preprint* 1/1993, Physics Department, Moscow State University, pp 1–5; 1993 *Zh. Eksp. Teor. Fiz.* **103** 1–10 (Engl. transl. 1993 *Sov. Phys.–JETP* **76** 1085–9)
[21] Lindsay S M, Burgess S and Shepherd I W 1977 *Appl. Opt.* **16** 1404–7