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FOLDED AND UNFOLDED ACOUSTIC MODES IN LONG-PERIOD GaAs-AlAs SUPERLATTICES. A COMBINED RAMAN-BRILLOUIN STUDY

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Résumé : Pour la première fois on observe simultanément les modes acoustiques directs (raies Brillouin) ainsi que les modes acoustiques repliés à des fréquences inférieures et supérieures aux raies Brillouin. Des courbes de dispersion très précises sont obtenues pour ces modes, à la fois pour les polarisations LA et TA et pour des vecteurs d'onde d'excitation dans la première et la seconde zone de Brillouin. Les intensités relatives des modes acoustiques repliés et des modes acoustiques directs sont mesurées et comparées à une théorie améliorée de l'interaction acousto-optique. L'accord est satisfaisant et permet une détermination précise du rapport p'_{13}/p_{13} des constantes photoélastiques de AlAs et GaAs.

Abstract : Direct acoustic modes (Brillouin lines) as well as folded acoustic modes at frequencies higher and lower than the Brillouin lines are simultaneously observed. Both LA and TA polarizations are investigated and very precise dispersion curves are obtained for values of the excitation wavevectors lying in the first and second Brillouin zone. The relative intensities of folded and direct acoustic modes are measured and compared to an improved theory of acousto-optic interaction. The agreement is good and allow a precise determination of the ratio p'_{13}/p_{13} of the photoelastic constants in AlAs and GaAs.

In the process of light scattering by acoustic phonons propagating normally to the layers of a superlattice, the wavevector k of the involved phonons is

given by $k = q + 2\pi m/D$, with $q = k_s - k_i$ (for anti-Stokes process). Here D is the period, q is the excitation wavevector; k_i and k_s are the wavevectors of incident light and scattered light respectively. The integer m is a folding index which labels the different modes ($m = 0$ corresponds to the Brillouin line).

Till now, only phonons corresponding to excitation wavevectors q less than the limit π/D of the first Brillouin zone have been probed in GaAs-AlAs superlattices. Here we investigate superlattices whose period D (in the range 200 ~ 500 Å) allows phonon modes observation in both sides of the boundary π/D . The variation of q are experimentally obtained by changing the wave-length λ of the incident light ($q = 4\pi n/\lambda$ for backward scattering experiments, n being the effective refractive index in the superlattice which is also λ dependent).

A suitable experiment set-up combining the advantages of both Brillouin and Raman techniques have allowed a systematical study of LA, TA, FLA (folded longitudinal acoustic) and FTA (folded transverse acoustic) modes. All these modes, except the FLA [1-3], are studied for the first time. Frequencies as low as 1 cm^{-1} have been investigated. Very precise phonon dispersion curves have been obtained by using several excitation wavelengths belonging to the Krypton and Argon-ion lasers, and by investigating several superlattices of varying periods but with the same aluminium concentration. The phonon branches are plotted for excitation wavevectors in the first and second Brillouin zone (Fig. 1) in a standard form using the normalized parameters $\Omega = \omega D/2\pi c$ and $Q = qD/\pi$, which are dimensionless (c is the light velocity in vacuum). One of the most striking effects in the case of $Q > 1$ is the observation of folded acoustic modes at frequencies below the direct acoustic mode (Fig. 2.a). The relative frequency positions of the folded modes are also changed (Fig. 2.b).

The intensity behaviour of the different modes depends strongly on Q . For Q values closed to 1, the intensity of FLA_1 can even be larger than that of LA (Fig. 3). Several models [1,3,4] have been proposed to calculate the intensities. We have used an improved new model which considers the refractive index modulation in the superlattice, as well as the modulation of elastic and photoelastic properties. The intensities of LA and FLA are calculated in different Brillouin zones (the details of our model and its comparisons with previous ones will be presented elsewhere). All the experimental results agree well with the theoretical predictions. In table I we present the comparisons between theory and experiments for the lowest frequency modes in two long-period superlattice samples.

Table I : Frequency and intensity of LA and FLA modes in two GaAs-AlAs superlattices. The Brillouin line is taken as the intensity reference.
 $\lambda = 4880 \text{ \AA}$; $n = 4.375$; $n' = 3.37$; p'_{13}/p_{13} is taken equal to 0.43 in order to obtain the best fit for the intensities.

MODES	$d = 104 \text{ \AA}, d' = 369 \text{ \AA} (Q = 1.41)$				$d = 114 \text{ \AA}, d' = 307 \text{ \AA} (Q = 1.27)$			
	frequency (cm^{-1})		intensity		frequency (cm^{-1})		intensity	
	calculated	measured	calculated	measured	calculated	measured	calculated	measured
FLA ₋₁	1.13	1.20	0.289	0.30	1.54	1.58	0.239	0.22
LA	2.67	2.68	1	1	2.69	2.68	1	1
FLA ₋₂	4.94	4.71	0.201	0.23	5.79	5.60	0.143	0.17
FLA ₊₁	6.48	6.2	0.303	0.30	6.93	6.7	0.284	0.30
FLA ₋₃	8.73	8.3	0.067	0.04	10.01	9.8	0.015	0.018
FLA ₊₂	10.28	9.9	0.123	0.12	11.18	10.8	0.072	0.09

It can be shown that the intensity ratios FLA_m/LA ($m = \pm 1, \pm 2, \dots$) are very sensible to p'_{13}/p_{13} (p'_{13} and p_{13} are the photoelastic constants (*) of AlAs and GaAs respectively), contrary to $\text{FLA}_m/\text{FLA}_{m'}$ ($m' = \pm 1, \pm 2, \dots$). Thus the intensity measurement of the folded modes with respect to the Brillouin line is necessary for the precise determination of p'_{13}/p_{13} . The calculated intensities in table I are obtained by taking p'_{13}/p_{13} equal to 0.43 for $\lambda = 4880 \text{ \AA}$, which gives the best fit between the theoretical and experimental results. Similarly we obtained $p'_{13}/p_{13} = 0.66$ for $\lambda = 4579 \text{ \AA}$ and $p'_{13}/p_{13} = 0.23$ for $\lambda = 6764 \text{ \AA}$.

As to the FTA modes, which are due to a polarization leakage [6], one finds that they are not polarized, contrary to the FLA. The intensities of the FTA modes are actually much weaker, but in the geometry of crossed polarizations for incident and scattered light, the FTA modes can be clearly seen (Fig. 4), contrary to the FLA which are then Raman inactive.

(*) According to Ref. 5, a longitudinal strain S_z induces a variation $\delta\epsilon_x$ of the dielectric constant given by $\delta\epsilon_x = -\epsilon_x^2 p_{13} S_z$, where p_{13} is the appropriate Pockels photoelastic constant.

Fig 1- Standard dispersion curves for GaAs-AlAs with a mean aluminium concentration $x=0.74$

x longitudinal • transverse (experimental)
Full and dashed lines are calculated from the elastic model.

Fig 4- The FLA₁ peak becomes more intense than the Brillouin LA ($Q = .78$; $x = .45$).

Fig 2- Folded longitudinal acoustic modes below (a) and above (b) the Brillouin lines ($Q = 1.32$; $x = .74$)

Fig 5- Folded transverse acoustic modes ($x = .74$; $Q = 0.52$) for crossed polarizations of the incident and scattered light.

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