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RESUME - Nous présentons la première détermination, à partir de la diffusion Raman, d'une grande partie des courbes de dispersion des modes acoustiques repliés dans des super-réseaux GaAs/GaAlAs de grandes périodes, ainsi que la première observation sans ambiguïté d'un mode optique replié. L'analyse des courbes de dispersion acoustiques fournit des informations sur les vibrations de l'alliage. Un modèle du type Kronig-Penney est présenté pour décrire l'ensemble des courbes de dispersion des phonons.

ABSTRACT - We report the first Raman scattering determination of large parts of the dispersion curves of folded acoustical modes in large period GaAs/GaAlAs superlattices and the first unambiguous observation of a folded optical mode. The analysis of the acoustical dispersion curves gives informations on the lattice dynamics of the alloy. A Kronig-Penney type model of the whole phonon dispersion curves of a superlattice is presented.

In this paper,we present new experimental results on lattice dynamics of GaAs/$Ga_{1-x}Al_xAs$ superlattices. The new periodicity in the growth direction induces,in superlattices,new modes in the Brillouin zone center,which should imply the onset of new Raman lines. Up to now the following observations have been reported: i) a low energy line unambiguously attributed /1/ to the first folding of the longitudinal acoustical (LA) mode,ii) an additional line a few cm-1 below the longitudinal optical (LO) line of GaAs,which has been assigned either to a folded LO phonon /2/ or to a zone center LO mode propagating parallel to the layers /3/. Both results have been obtained in resonant conditions. We report here the observation in non resonant conditions of folded acoustical and optical Raman lines and analyse the anomalous incident wavelength dependence of the former,in large period samples, as due to the phonon dispersion. We thus demonstrate that one can obtain in large period superlattices,by means of Raman backscattering, i) large parts of the phonon dispersion curves (PDC) of the superlattices and ii) useful informations on the PDC of the bulk constituents,which could hardly been determined by neutron scattering. Theoretical predictions of the whole PDC for longitudinal modes propagating along the growth direction,using a Kronig-Penney type model,are presented.

The studied samples have been grown by Molecular Beam Epitaxy on (100) oriented GaAs substrates and characterized by X-ray diffraction. Sample A: thickness of the GaAs layer d_1= 56 Å,of the $Ga_{1-x}Al_xAs$ layer d_2=78 Å with x= 0.77; Sample B : d_1=150Å d_2= 107 Å,x = 0.36; Sample C : $d_1=d_2$= 22.6 Å and x= 0.3. /4/

Concerning the acoustical range,we have observed by backscattering on sample A and B at room temperature low energy lines for all Krypton and Argon ion laser incident wavelength (up to five lines on Sample B at 6764A). Three low energy spectra obtai-

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ned on Sample A are drawn on Fig.1: the four lines are assigned to the four first folded LA branches (see definition on Fig.2). The high intensity of these lines, which allows their observation in non resonant conditions, is mainly due to their very small frequency shift and thereby to the high value of the related thermal Bose factor. Let us outline the variation of the frequency shifts with incident laser energy. Sample A and B displaying a large superperiod, the Brillouin zone edge wavevector is small (2.34 10^6 cm^{-1} for Sample A and 1.22 10^6 cm^{-1} for Sample B) and is thus of the same order of magnitude as the created phonon wave vector q. Assuming the refraction index to be constant and equal to 4. , q varies between 1.05 10^6 cm^{-1} at 4765 Å and 0.74 10^6 cm^{-1} at 6764 Å. The Raman lines must therefore be assigned to non zone center phonons and the change of the frequency shift with

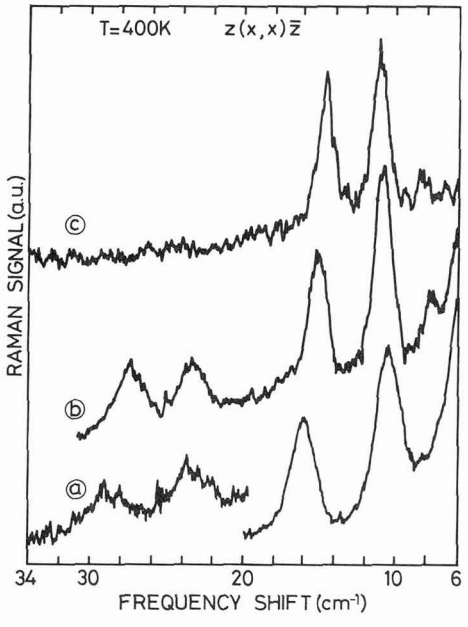

Figure 1: Raman scattering spectra on Sample A for three different wavelength: a)4765Å ,b)5682Å ,c)6764Å.

incident energy reflects the dispersion curves. Figure 2 compares the experimental PDC with theoretical ones obtained from the elastic model /5/:

$$\cos(q(d_1+d_2)) = \cos\frac{\omega d_1}{v_1}\cos\frac{\omega d_2}{v_2} - \alpha\sin\frac{\omega d_1}{v_1}\sin\frac{\omega d_2}{v_2}$$

with:

$$\alpha = \frac{(\rho_1 v_1)^2+(\rho_2 v_2)^2}{2\rho_1 v_1 \rho_2 v_2}$$

where ρ_i, v_i are density and sound velocity in medium i. Using the experimental sound velocity of GaAs /6/ and the theore-

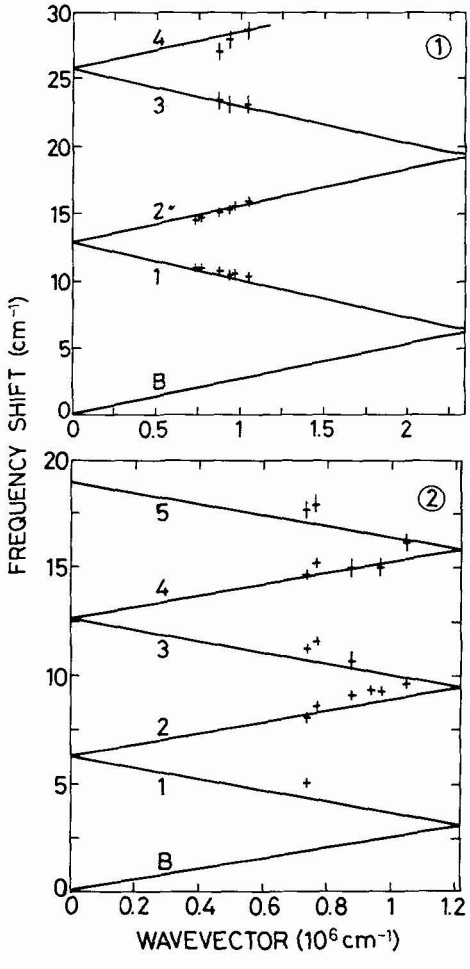

Figure 2: Experimental results on Sample A (1) and Sample B (2) compared with the elastic model. The different branches are indexed by B (Brillouin line) or their "folding number".

tical one of AlAs /7/ and a virtual crystal approximation for $Ga_{1-x}Al_xAs$, a good fit is obtained without any adjustable parameter.

The determination of the dispersion curves of the superlattice allows to test the validity of the lattice dynamics models of the constituents: we obtain here some information on the lattice dynamics of the alloy. The sharpness of the folded lines, together with the fit between experiment and predictions of the elastic model demonstrate that the low energy alloy vibrations are fairly well accounted for within VCA. Besides, Sample A displays, for an incident energy close to the $E_0(Ga_{1-x}Al_xAs)$ gap, low energy structures identical to those already observed on same composition GaAlAs monolayer. We assign in both cases these structures /8,9/ to disorder induced zone edge acoustical phonons (DATA, DALA). This coexistence in the alloy of dispersive VCA and non dispersive disorder induced acoustical contributions is actually predicted in a Coherent Potential Approximation model of the GaAlAs lattice dynamics : using the simplest model /10/ (linear chain with nearest neighbour coupling), which discards the transverse properties, we calculated the spectral density of states (SD) of $Ga_{.23}Al_{.77}As$ for different values of q. In the acoustical range (see Figure 3) for a given q, the SD can be split up into two parts:

i) a sharp intense and dispersive one whose maximum position identifies with the VCA acoustical mode one. The latter, which gives rise to the folded lines, is weakly affected by the disorder and can thus be treated as an elastic wave.

ii) a broad weak non dispersive one, disorder induced, which maximum intensity is located at a frequency close to the VCA zone edge acoustical mode one. It corresponds to the DALA "mode". One can thus describe the dispersion of the acoustical modes in the mixed crystal by drawing two "thick curves" /8/ corresponding to both contributions, as can be seen in the insert of Figure 3.

Beside the GaAs LO mode, the two GaAlAs LO modes (GaAs-type and AlAs-type) and two folded acoustical lines, Sample C displays an additional line about 5 cm^{-1} below the GaAs LO one, which is well resolved at liquid Helium temperature (see Fig.4). The observation of this new optical line has been done in $z(x,y)\bar{z}$ configuration, using a 5145Å incident wavelength i.e. clearly out of resonance. We can therefore rule out an assignement to a LO mode propagating parallel to the layers plane. Such a symmetry forbidden mode could only appear in the Raman signal, due to the Fröhlich interaction, in resonant conditions, in the $z(x,x)\bar{z}$ configuration /3/. We assign therefore our additional line to a folded LO mode. This is, to our knowledge, the first unambiguous report of a folded optical mode in a superlattice. A detailed study of this mode is planed and require the development of a whole lattice dynamics model for large period superlattices.

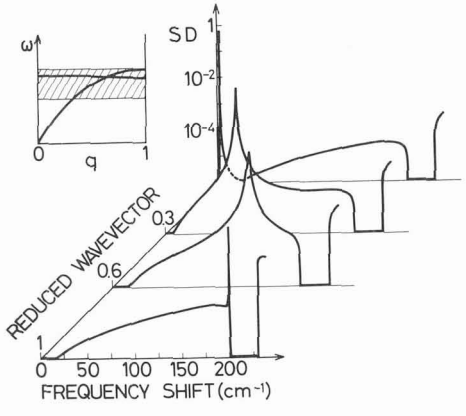

Figure 3: Spectral density of states (SD) of $Ga_{.23}Al_{.77}As$ calculated for four values of q: 1%, 30%, 60% and 99% of the Brillouin zone edge wavevector. The energy range has been truncated at the beginning of the optical band. The insert is a schematic representation of the thick dispersion curves in the acoustical range. The hatched region delimits the width of the DALA mode. The acoustical mode one is negligible at the figure scale.

We present here a Kronig-Penney type model of the dispersion curves along the superlattice axis: for a given frequency, the eigenmodes of the superlattice are assumed to reduce,in each layer,to a linear combination of a backward and a forward propagating eigenmodes of the corresponding bulk material. This assumption is only

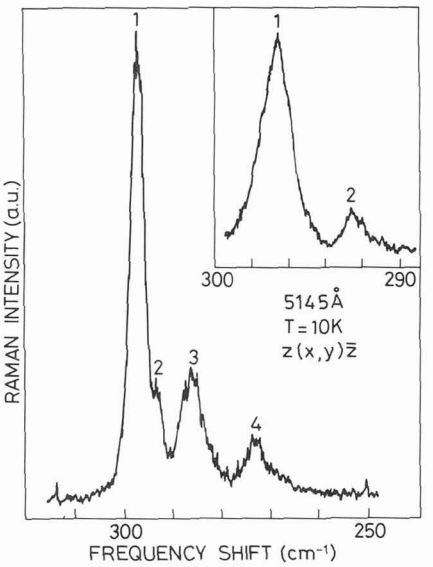

Figure 4: Raman scattering spectrum on Sample C. The detail in the insert has been obtained at higher resolution. The following labels are used:
1: LO line of GaAs, 2: folded LO mode
3: GaAs-type LO mode of GaAlAs
4: leakage of the TO mode of GaAs

valid for not too small layer thicknesses.The superlattice dispersion curves are obtained by writing the superperiodicity of the eigenmodes and some boundary conditions. In the case of a superlattice designed with two binary compounds AB and AC, the interface is build up with A atoms belonging to both compounds. The boundary conditions then reduce to the identification of the displacement of the interface A atom with the ones it would have,embedded in AB or in AC. Should one use linear chain models to describe longitudinal modes in both compounds,the dispersion curves at low frequency reduce to those predicted by the elastic model. Details will be published elsewhere. To give a feeling of the results,let us assume here the same nearest neighbour force constants and the same lattice spacing a for both materials,which is a good approximation to describe the GaAs/AlAs system. The dispersion curves are then the solution of:

$$\cos(q(d_1+d_2)) = \cos(k_1 d_1)\cos(k_2 d_2) - \alpha \sin(k_1 d_1)\sin(k_2 d_2)$$

with:

$$\alpha = \frac{1-\cos(k_1 a)\cos(k_2 a)}{\sin(k_1 a)\sin(k_2 a)}$$

k_i being the (eventually complex) wavevector,at frequency ω,in medium i.

As we assume here the heaviest atom to be common to both compounds (like As in GaAs and AlAs),the acoustical frequency range coincide in both materials. Thus,for any acoustical frequency, propagating modes exist in both layers and build up folded dispersive delocalized modes in the superlattice. On the contrary,in the optical range,the frequency bands of the bulk materials can be partially overlapping or even well separated. In the latter case,the vibrations are localized in one of the two layers and displays thereby no dispersion: the superlattice behaves like a phonon "multi-quantum well" (see Fig.5 a). If the bulk optical bands are partially overlapping, dispersive delocalized modes only appear in the common frequency range (see Fig. 5 b).

This model cannot be directly used in the case of GaAs/$Ga_{1-x}Al_xAs$ superlattice as the alloy exhibits a two mode behaviour (GaAs and AlAs type bands). One can however draw some qualitative predictions:
i) the AlAs-type vibrations should be localized in the alloy layer (like band 2 in Fig.5 a).

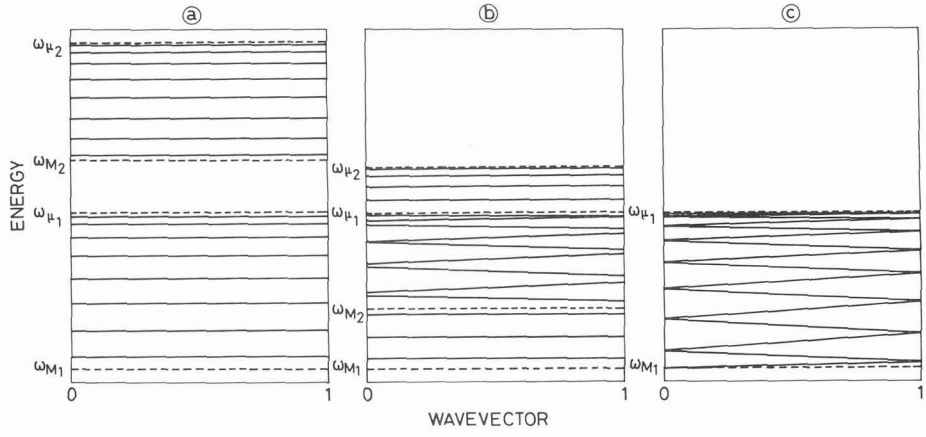

Figure 5: Optical dispersion curves predicted by the Kronig-Penney model for a linear chain of alternating 8 unit cells of medium 1 (M_1=70amu,m_1=50amu) and 8 unit cells of medium 2 (M_2=M_1=70amu and m_2= a)25amu,b)40amu,c)50amu). μ is the reduced mass.

ii) the GaAs-type vibrations in GaAlAs should partially interact with the optical vibrations of pure GaAs.
iii) the zone center optical frequency of pure GaAs being greater than the GaAs-type one in $Ga_{1-x}Al_xAs$,Lines 1 and 2 (see Fig.4) should correspond to non dispersive modes localized in the GaAs layers.

Calculations are in progress to generalize this model to superlattices containing alloys.

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