PROC. 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980 J. PHYS. SOC. JAPAN **49** (1980) SUPPL. A p. 1021–1023

PHONON FOLDING AND ANISOTROPY IN GaAs-Alas SUPERLATTICES

R. Merlin,* C. Colvard, M.V. Klein, H. Morkoç

University of Illinois at Urbana-Champaign Urbana, IL 61801, USA

A. C. Gossard and A. Y. Cho Bell Laboratories, Murray Hill, New Jersey 07974, USA

Raman scattering has been used to study two distinctive features of the lattice dynamics of a superlattice: the folding of the Brillouin zone which results in the appearance of gaps in the phonon spectrum for wavevectors satisfying the Bragg condition, and the anisotropy of polar phonons induced by layering.

The properties of the phonons in GaAs-Ga_{1-x}Al_xAs superlattices have recently received much attention [1-4]. The phonon spectrum of these materials shows two interesting features which are dependent on the layer thicknesses. First, the new periodicity along the direction perpendicular to the layers results in a smaller, folded Brillouin zone (BZ) and the appearance of gaps in the phonon dispersion for wavevectors satisfying the Bragg condition [1,2,4]. Second, the layering gives rise to anisotropic effects which may be absent in the constituent materials, as is the case for superlattices made out of Ga_{1-x}Al_xAs [3].

Phonons in semiconductor superlattices have been studied by means of infrared spectroscopy by Barker et al. [1] and through the use of tunnelling techniques by Narayanamurti et al. [2]. Results of Raman scattering (RS) studies on superlattices have been reported by us [3,4] and other groups [1,5,6]. A summary of our results will be given here.



Figure 1 Raman spectrum of the 14 Å GaAs - 11 Å AlAs superlattice

Both effects, phonon folding and anisotropy, appear in the Raman spectrum shown in Fig. (1), of a superlattice made of alternating layers of 14 Å GaAs and 11 Å AlAs. This sample was grown by molecular beam epitaxy (MBE) on a GaAs substrate oriented such that the layers are perpendicular to the z = [001] direction. Only longitudinal modes are allowed for a Raman backscattering experiment from a {001} face. The two narrow lines at 63.1 and 66.9 cm⁻¹ correspond to the scattering from longitudinal acoustic (LA) phonons folded to the zone center from $q = 2\pi/d$ in the extended zone, where d is the superlattice period. The peaks at 277 and 288 cm⁻¹ derive from the longitudinal optic (LO) phonon of bulk GaAs. The layering gives rise to the splitting of this phonon, which becomes an E - or a B₂ - symmetry mode when it propagates parallel or perpendicular to the layers [3]. The LO - phonon of AlAs splits giving the $E(LO_2)$ and $B_2(LO_2)$ modes which appear in the spectrum of Fig. (1) at 382 and 399 cm⁻¹ [3].



 ≥ 2 Calculated phonon dispersion for the 14 Å GaAs - 11 Å AlAs superlattice [4]: The dots are the experimental values of the frequencies of the q $\sim 2\pi/d$ folded IA phonons

Lattice dynamic calculations using a linear chain model have been reported for longitudinal modes by Barker et al. [1] and for transverse phonons by Tsu and Jha [7]. We have taken a different approach by considering the low lying phonons to be well represented by elastic waves in a layered continuum [4]. We have calculated the phonon dispersion for the 14 Å - 11 Å sample using only the experimental values of the elastic constants of bulk GaAs and AlAs [4]. Figure (2) shows the calculated phonon dispersion in the frequency region where the first folding occurs. The layering induces a splitting of LA phonons at $q = \pm 2n\pi/d$ into modes with A_1 and B_2 symmetry in the tetragonal superlattice [1]. As shown in Fig. (2), the experimental values for the $A_1^{(1)}$ and $B_2^{(1)}$ phonon frequencies agree very well with the calculation.

We now briefly discuss the microscopic mechanisms involved in the scattering from folded LA phonons. First, there is a contribution which originates in the deformation potential electron-phonon interaction. Another term, intrinsic to layered materials, derives from the phonon-modulation of the layer thicknesses [4]. Both interactions contribute to the scattering from the even-parity A⁽¹⁾mode but give a vanishing contribution to the odd-parity B⁽¹⁾-scattering [4]. The B⁽¹⁾-phonon is forbidden in the $z(xx)\overline{z}$ configuration by the usual RS selection rules. This "forbidden" B⁽¹⁾-scattering, as in the case of the well known "forbidden" LO(Γ)-scattering, originates in the Fröhlich interaction between electrons and the macroscopic longitudinal electric field carried by this mode [4].

The folded LA phonons show a strong resonant enhancement for laser energies near the fundamental gap of the 14 Å - 11 Å superlattice at about 1.96 eV [4]. Based on the mechanisms sketched above and using the Kronig-Penney model for the electrons, we have calculated the energy-dependence of the $A_1^{(1)}$ and $B_2^{(1)}$ scattering cross sections [4]. The results of this calculation show good agreement with the resonance data and are reported elsewhere [4].

We now consider the layering-induced splitting of LO-phonons. We have observed this effect in the 14 Å - 11 Å sample and also in a superlattice with periods of 50 Å GaAs-50 Å AlAs [3]. The LO modes carry a macroscopic longitudinal electric field. The anisotropy originates in the different boundary conditions for fields parallel and perpendicular to the layers. For the B_2 -modes, which propagate along z, the electric field is confined in one of the two media, whereas the field associated with an E(LO) phonon is uniform over the layers. The boundary conditions at the interfaces give rise also to anisotropic dielectric properties. For thin layer superlattices, the material behaves like a uniaxial crystal whose dielectric constants are given by [8].

 $\epsilon_{x,y} = (d_1 \epsilon_1 + d_2 \epsilon_2)/d$, $\epsilon_z = \epsilon_1 \epsilon_2 d/(d_1 \epsilon_2 + d_2 \epsilon_1)$,

where ϵ_1 and ϵ_2 are the bulk dielectric constants of the two layers, with thicknesses d_1 and d_2 . The frequencies of the LO phonons of B_2 and E symmetry are given by the zeros of the frequency-dependent ϵ_z and $\epsilon_{x,y}$. The frequencies of the $B_2(LO)$ modes coincide with those for the bulk materials. Using the expression for $\epsilon_{x,y}$ and the optical constants from the literature the frequencies of the E(LO) modes can be determined. The calculated frequencies [3] show good agreement with our data, as well with data previously published on GaAs-Ga_{0.75} Al_{0.25}As superlattice [6] in which the peaks we assign to E(LO) scattering are ascribed to electronic umklapp processes.

Finally, some remarks about wavevector conservation. The E(LO) modes seen in Fig. (1) are forbidden in the $z(\cdots)\overline{z}$ geometry. This scattering is induced by the Fröhlich interaction and most likely originates in the presence of defects or disorder, which induces the breakdown in wavevector conservation [3]. A similar effect has been observed in other polar semiconductors, particularly CdS [9], where it was shown that a very small amount of impurities lead to spectra very similar to ours in Fig. (1).

*Present address: Department of Physics, University of Michigan, Ann Arbor, Michigan 48109.

- 1) A.S. Barker, Jr., J.L. Merz and A.C Gossard: Phys. Rev. B17 (1978) 3181.
- V. Narayanamurti, H.L. Störmer, M.A. Chin, A.C. Gossard and W. Wiegmann: Phys. Rev. Lett. 43 (1980) 1536.
- R. Merlin, C. Colvard, M.V. Klein, H. Morkoç, A.Y. Cho and A.C. Gossard: Appl. Phys. Lett. 36 (1980) 43.
- 4) C. Colvard, R. Merlin, M.V. Klein and A.C. Gossard: Phys. Rev. Lett. 45 (1980) 298.
- P. Manuel, G.A. Sai-Halasz, L.L. Chang, Chin-An Gang and L. Esaki: Phys. Rev. Lett. 37 (1976) 1701.
- 6) G.A. Sai-Halasz, A. Pinczuk, P.Y. Yu and L. Esaki: Solid State Commun. 25 (1978) 381.
- 7) R. Tsu and S.S. Jha: Appl. Phys. Lett. 20 (1972) 16.
- 8) S.M. Rytov: Sov. Phys. JETP 2 (1956) 466.
- 9) S. Permogorov and A. Reznitsky: Solid State Commun. 18 (1976) 781.