Observation of Folded Acoustic Phonons in a Semiconductor Superlattice*

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(Received 9 May 1980)

Raman scattering from folded longitudinal phonons in a GaAs-AlAs superlattice is observed, and the resonant behavior for laser frequencies near the gap of this sample is studied. A quantitative explanation of the data is given in terms of a simple theory which involves the Kronig-Penney model for the electrons and considers the phonons in the elastic continuum limit.

PACS numbers: 78.30.Gt, 63.20.Dj, 68.55.+b, 78.65.Jd

In a superlattice the new periodicity along the direction perpendicular to the layers is expected to result in Brillouin-zone (BZ) folding and the appearance of gaps in the phonon spectrum for wave vectors satisfying the Bragg condition. Recently Narayanamurti *et al.*[1] used tunneling techniques to observe this effect for phonons at $q = \pi/d$, where d is the superlattice period. Phonons at $q = 2n\pi/d$, i.e., at the center of the folded BZ, are in general accessible by means of light scattering, and were reported in Raman-scattering (RS) studies of (GaAs, AlAs) superlattices.[2,3] However, it was later shown that the features attributed to these phonons were likely due to insufficient corrections for scattering from air at the sample surface.[4]

In this Letter we report the first clear evidence of RS from folded acoustic phonons in a superlattice.[5] We show that the Raman data can be explained by a theory which treats the phonons in the elastic continuum limit, considers a Kronig-Penney model for the electrons, and uses a modified Fröhlich coupling and a novel deformation-type electron-phonon coupling.[6]

The sample investigated in this work consists of ~1720 periods of 13.6 Å GaAs–11.4 Å AlAs and was grown by molecular-beam epitaxy on a GaAs substrate with the layers perpendicular to the $z = [001]$ direction.

Figure 1 shows the Raman spectrum of the superlattice for laser energy $\omega_L = 1.959$ eV in the $z(x,x)\bar{z}$ backscattering configuration at $T = 300$ K. The features for shifts $\Omega > 200$ cm^{-1} are discussed elsewhere.[4] The spectrum shows two new peaks at 63.1 and 66.9 cm^{-1} and a broad structure at $\Omega \sim 100$ cm^{-1}. Transverse phonons are forbidden in backscattering from a {001} face. We assign the two narrow lines to scattering from longitudinal acoustical (LA) phonons folded to the zone center from $q = 2\pi/d$ in the extended zone. The basis for this assignment is provided by the calculated pho-

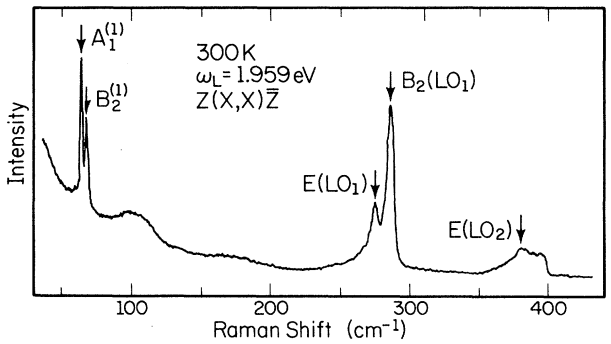

FIG. 1. Raman spectrum of the superlattice.

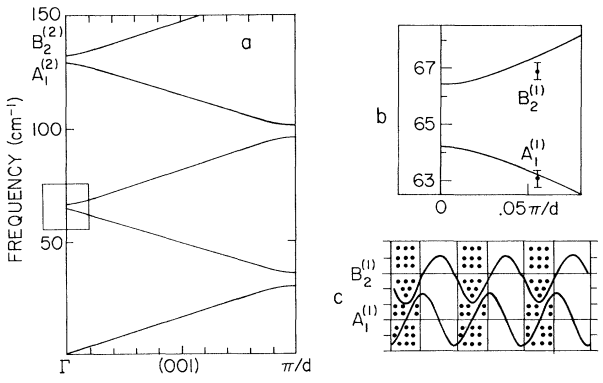

FIG. 2. (a) Dispersion of longitudinal phonons in the superlattice. The region indicated by a square is shown in detail in (b), where the dots correspond to the experimental values for the phonon frequencies. (c) Amplitude of the folded phonons. The dotted regions correspond to AlAs layers.

non dispersion shown in Fig. 2 to be discussed next.

Barker, Merz, and Gossard[3] have used a linear-chain model to calculate the phonon dispersion in small layer superlattices. We have taken a different approach by considering the low-lying phonons to be well represented by elastic waves in a layered continuum. In the continuum limit the Ω versus q relation for longitudinal vibrations in a layered structure with layer thicknesses d_1 and d_2 is given by[7]

$$\cos(qd) = \cos\left(\frac{\Omega d_1}{c_1}\right)\cos\left(\frac{\Omega d_2}{c_2}\right) - \frac{1+\kappa^2}{2\kappa}\sin\left(\frac{\Omega d_1}{c_1}\right)\sin\left(\frac{\Omega d_2}{c_2}\right), \quad (1)$$

where $\kappa = c_1\rho_1/c_2\rho_2$, c_1 and c_2 are the sound velocities for LA phonons along [001] for both media, and ρ_1 and ρ_2 are the corresponding densities. The phonon dispersion for our sample is shown in Fig. 2(a). We have used for[8] GaAs $\rho_1 = 5.3149$ g/cm^3 and $c_1 = 4.726\times10^5$ cm/sec and for[9] AlAs $\rho_2 = 3.7285$ g/cm^3 and $c_2 = 5.118\times10^5$ cm/sec. The layering gives rise to a splitting of longitudinal modes with $q = \pm 2n\pi/d$ into phonons with A_1 and B_2 symmetry in the tetragonal superlattice.[3] As is shown in Fig. 2(b), the positions of the two new lines in the spectrum of Fig. 1 agree very well with the calculated frequencies for the $A_1^{(1)}$ and $B_2^{(1)}$ phonon. The phonon dispersion calculation also suggests a tentative assignment for the broad structure at $\Omega \sim 100$ cm^{-1}, as due to disorder-induced RS from longitudinal modes at the border of the folded BZ.

The $A_1^{(1)}$ and $B_2^{(1)}$ lines in Fig. 1 show a strong resonance enhancement at $\omega_L \sim 1.96$ eV which corresponds roughly to the value of the energy gap of our sample[10] (see Fig. 3). Indeed the lines are only seen close to resonance. The enhancement is larger for the line labeled as $B_2^{(1)}$ as shown by the dip in the data of Fig. 3(b).

To understand the coupling of light to these two phonons we apply the usual theory of resonance RS to virtual valence- to conduction-band transitions of carriers in the superlattice. We use the Kronig-Penney model and assume the potential discontinuities for the conduction and valence band to be $0.85\Delta V_g$ and $0.15\Delta V_g$,[11] where ΔV_g is the difference between the E_0 gaps of AlAs and GaAs. The electron, heavy-hole, and light-hole masses are assumed to be as in GaAs: $0.0665m_e$, $0.45m_e$, and $0.08m_e$, respectively. We find the heavy hole to conduction band and light hole to conduction band edges at $E_{hh} = 2.012$ eV and $E_{lh} = 0.027$ eV. Because of its proximity with these calculated values, we assign the peak in the resonant RS spectrum in Fig. 3(a) to these transitions. The heavy- and light-hole resonances are not resolved in the data, probably because of damping effects. Using the values indicated above and the Kronig-Penney model we find $0.073m_e$, $0.513m_e$, and $0.080m_e$ for the electron, heavy-hole, and light-hole masses along the z direction. These values imply that we have a three-dimensional (3D) situation, i.e., the electrons are not confined to the wells as in the 2D resonant Raman studies previously reported.[12]

The coupling of the $A_1^{(1)}$ phonon to these electrons was calculated by considering the phonon modulation of the layer thicknesses, and the stress-induced bending of the potential wells.[6] The former mechanism involves no adjustable parameters; the latter involves the usual deformation potentials of GaAs and AlAs. This essen-

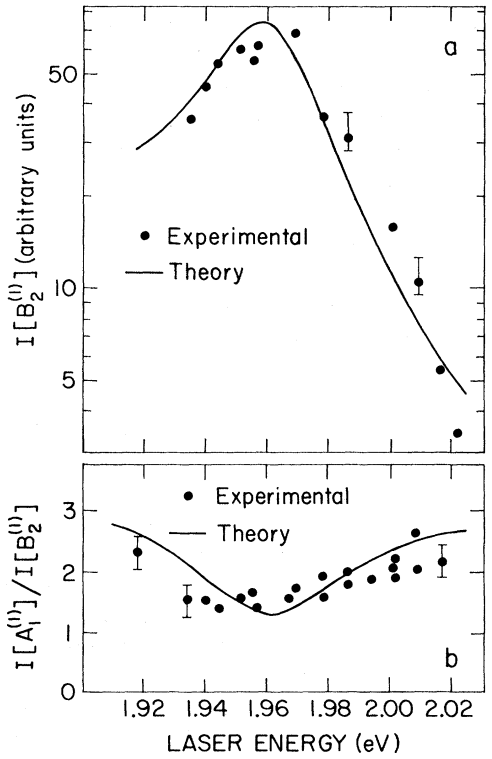

FIG. 3. (a) Resonant enhancement of the $B_2^{(1)}$ mode; (b) intensity ratio as a function of laser energy.

tially scalar coupling mechanism gives the (x,x) selection rule seen for the $A_1^{(1)}$ phonon in Fig. 1.

We observed neither peak in the $z(x,y)\bar{z}$ geometry which allows scattering from B_2 phonons.[13] Within the Kronig-Penney model the absence of *allowed* $B_2^{(1)}$ scattering results from the inversion symmetry of the model, since B_2 modes transform like z in the D_{2d} point group of the superlattice.

The $B_2^{(1)}$ mode is forbidden in the $z(x,x)\bar{z}$ geometry by the usual Raman selection rules.[13] Under resonant conditions forbidden LO(Γ) RS induced by the Fröhlich interaction has been observed in bulk GaAs.[14,15] Like the LO(Γ), the LA phonons along [001] may carry a longitudinal electric field through terms involving the dispersion of the piezoelectric tensor. Therefore, forbidden RS from folded LA phonons may also take place[16] in analogy with the forbidden LO scattering. We assign the $B_2^{(1)}$ line in Fig. 1 to this type of scattering.

We have made appropriate modifications to the usual theory[17] of the Frölich interaction to describe both "normal" and "umklapp" intraband scattering of carriers with emission of $B_2^{(1)}$ phonons.[6] The effective charge of these phonons is a free parameter in the theory.

Using standard expressions,[18] we have calculated the ω_L dependence of the Raman tensor for the folded phonons, with the electronic wave functions derived from the Kronig-Penney model. The experimental points in Fig. 3(a) are not corrected for the penetration depth of the light. The calculated curve is $|R_b|^2/\sigma(\omega_L)$, where R_b is the Raman tensor and $\sigma(\omega_L)$ is the absorption coefficient. This curve has been shifted to lower energies by 50 meV and moreover a damping factor of 20 meV has been included; $\sigma(\omega_L)$ was calculated through its proportionality to the density of states as in Ref. 12.

The calculated ω_L dependence for the intensity ratio is shown in Fig. 3(b). Because the amplitude of the A_1 modes is an odd function [Fig. 2(c)], the Fröhlich contribution to the $A_1^{(1)}$ scattering vanishes. The dip in the data originates in the wave-vector–dependent terms contributing to the forbidden $B_2^{(1)}$ scattering. These terms exhibit a larger resonant enhancement than the k-independent terms.[18] From the fit with the experimental data we obtain for the effective charge $\alpha = 0.61$. This value agrees within an order of magnitude with the value of $\alpha = 0.28$, which we obtain using the linear-chain model of Ref. 3.

We finally mention the lack of observed scattering from $q = 2n\pi/d$ folded LA phonons for $n > 1$. Using the theory outlined above, we find that the calculated scattering intensities for these phonons are more than two orders of magnitude smaller than the corresponding intensities for the $A_1^{(1)}$ or $B_2^{(1)}$ modes.

In summary, we have observed scattering from the folded LA phonons in a semiconductor superlattice. We have shown that the selection rules of the scattering and the resonant enhancement can be explained in terms of a simple theory involving the Kronig-Penney model for the electrons and by treating the folded phonons in the elastic continuum limit.

This work was supported in part by the Joint Electronics Program (U. S. Navy, U. S. Army, and U. S. Air Force) under Contract No. N00014-79-C-0424, the National Science Foundation under Grant No. DMR 7902780, and profited from institutional support by the National Science Foundation for the Materials Research Laboratory under Grant No. DMR 7723999.

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ERRATUM

HIGH-DENSITY EFFECTS ON THERMONUCLEAR IGNITION FOR INERTIALLY CONFINED FUSION. Stanley Skupsky [Phys. Rev. Lett. 44, 1760 (1980)].

In the paragraph before Eq. (3), $T^{3/2}/\eta_e$ should be $T^{3/2}/n_e$. Equation (3) should be

$$\lambda_R/\lambda_E \propto 1/n_e .$$

In the next set of equations,

$$\frac{1}{3}\frac{\partial}{\partial r}\varphi = -\frac{J}{\lambda_R}\left(\frac{JE}{\lambda_E}\right)$$

should read

$$\frac{1}{3}\frac{\partial}{\partial r}\varphi = -\frac{J}{\lambda_R} + \frac{\partial}{\partial E}\left(\frac{JE}{\lambda_E}\right).$$