PHYSICAL REVIEW B VOLUME 33, NUMBER 4 15 FEBRUARY 1986

Raman scattering study of acoustical zone-center gaps in GaAs/AlAs superlattices

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(Received 4 November 1985)

We present a detailed theoretical analysis of the folded acoustical gaps in superlattices within the framework of the elastic approximation and compare its predictions with both backward and forward Raman scattering experiments. We thereby obtain the first evidence of a zone-center gap between folded acoustical branches and also conclusive proof of the photoelastic nature of the involved light scattering process. A good quantitative description of the experimental results is obtained.

The folding of the acoustical dispersion curves in superlattices is well established:[1-4] Following the first observation by Raman backscattering[1] of folded longitudinal acoustical (LA) lines near zone center, it has been demonstrated[2] that one can determine parts of the folded LA dispersion curves by varying the incident laser frequency in a Raman backscattering experiment. These results are in very good agreement with the elastic model:[5] The folded LA dispersion curves are obtained first by folding the dispersion curve of an average bulk material and second by opening small gaps at zone center and zone edge. The Raman spectrum corresponding to the first folding appears then as a doublet whose splitting (see inset in Fig. 1, open circles) is due both to the gap opening at zone center and to dispersion. Indeed, the value of the wave vector of the phonon created during the backscattering experiment is never negligible relative to the Brillouin-zone extent. Consequently, the contribution of the dispersion to the splitting is always dominant. There is thus, in contrast with zone-edge gaps,[6] no experimental evidence of any gap opening at zone center. We present in this Rapid Communication the first evidence of this feature, in GaAs/AlAs superlattices, by means of forward Raman scattering: In this experiment the phonon wave vector is nearly exactly vanishing (see inset in Fig. 1, closed circles). Moreover, this configuration involves zone-center modes of well-defined tetragonal symmetry, thus providing a clear test of the role of the photoelastic process in the scattering mechanism.[3]

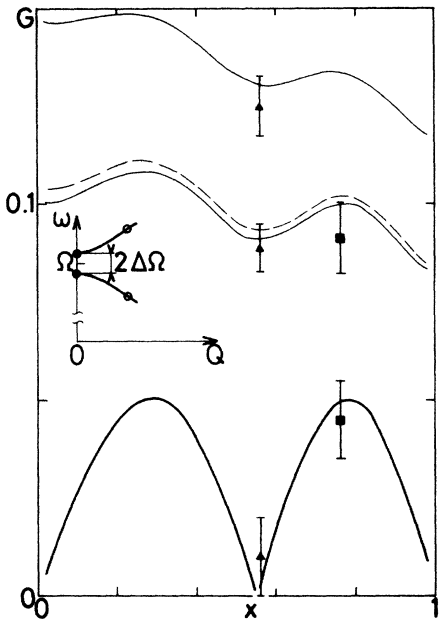

FIG. 1. Ratio of the first acoustical doublet splitting over its average frequency drawn as a function of the average aluminum concentration. The thick line corresponds to the zone-center calculation, which is independent of the period d. The thin lines correspond to the S1 period and two different phonon wave vectors related to a backscattering experiment done at 674 nm (lower curve) and 514 nm (upper curve). The dashed line corresponds to the S2 period and a backscattering experiment at 567 nm. The experimental results are represented by full triangles (S1) and full squares (S2).

Let us first derive the value of the gap at zone boundaries from the superlattice dispersion relation in the elastic approximation. This relation is written for a superlattice made of the periodic stacking of layers of thickness d_1 (d_2), sound velocity v_1 (v_2), and density ρ_1 (ρ_2):

$$\cos(Qd)=\cos\left(\omega\frac{d_1}{v_1}\right)\cos\left(\omega\frac{d_2}{v_2}\right)-\left(1+\frac{\alpha^2}{2}\right)\sin\left(\omega\frac{d_1}{v_1}\right)\sin\left(\omega\frac{d_2}{v_2}\right), \quad (1)$$

where $d=d_1+d_2$ is the period of the superlattice, Q is the superlattice wave vector at (angular) frequency ω, and where

$$\alpha=\frac{|\rho_1 v_1-\rho_2 v_2|}{(\rho_1 v_1 \rho_2 v_2)^{1/2}}$$

characterizes the superlattice modulation. Note that the acoustical impedances $\rho_i v_i$ ($i=1,2$) and not v_i are the relevant parameters. The modulation parameter α is usually small, and the superlattice dispersion relation reduces at zeroth order to

$$\cos(Qd)=\cos\left[\omega\left(\frac{d_1}{v_1}+\frac{d_2}{v_2}\right)\right].$$

This relation simply means that the transit time through a period is the sum of the transit time through both layers, a straightforward result when the reflectivity vanishes at each interface. One recognizes the folding of an average com-

pound of sound velocity v:

$$v=\frac{v_1 v_2}{x v_1+(1-x)v_2},$$

where x is the average concentration of compound 2 in the superlattice:

$$x=d_2/d \ .$$

The latter parameter provides, in conjunction with the period d, an alternative geometrical description of the sample, which will always be used in the following. Within this approximation, there is no gap opening, and the first folded zone-center energy is written as

$$\Omega=2\pi v/d \ .$$

Expanding up to second order the exact zone-center frequency ω as a function of $\Delta\Omega=\omega-\Omega$ leads to

$$\Delta\Omega^2=\alpha^2\frac{v^2}{d^2}\sin^2\pi\left[\frac{x v_1-(1-x)v_2}{x v_1+(1-x)v_2}\right] \ . \tag{2}$$

The gap opening ($2\Delta\Omega$) is inversely proportional to the period d. Let us therefore introduce the ratio G:

$$G=\frac{2\Delta\Omega}{\Omega}$$

$$=\frac{\alpha}{\pi}\left|\sin\pi\left[\frac{x v_1-(1-x)v_2}{x v_1+(1-x)v_2}\right]\right| \ . \tag{3}$$

This ratio, which expresses the width of the gap divided by the average energy, does not depend on d and vanishes at the concentration $x_c=v_2/(v_1+v_2)$. Its variation as a function of x is shown by the thick curve in Fig. 1, where we used the following parameters:

$$\rho_1=5.3149\ \text{g/cm}^3, \quad v_1=4.726\times10^5\ \text{cm/s (GaAs)} \ ,$$

$$\rho_2=3.7285\ \text{g/cm}^3, \quad v_2=5.75\times10^5\text{cm/s (AlAs)} \ .$$

ρ_1, ρ_2, and v_1 are standard values, whereas we determined v_2 by fitting the average energy Ω on a large number of GaAs/AlAs superlattices. Relation (3) fits very well the exact value of $\Delta\Omega$ computed from (1) and appears to be better than the other approximate relation given in Ref. 4 because of the significative coupling between the different folded modes.[7]

The maximum value of G amounts to about 0.05, which gives a gap of about 2 cm^{-1} for $d=4$ nm, a value which is larger than the usual Raman resolution (0.5 cm^{-1}). On the contrary, because of acoustical dispersion, the phonon energy becomes less dependent on the gap value as its wave vector increases. For a typical value of the wave vector of the phonon created by backscattering (10^6 cm^{-1}), the remaining energy modulation lies within the uncertainty range, as can be seen in Fig. 1. We plot in this figure the variation, as a function of x, of the ratio of the doublet splitting over the average frequency computed for some nonvanishing wave vectors. This corresponds to various backscattering incident wavelengths and to different periods. (This ratio is not independent of d as it is at zone center.)

We therefore performed forward-scattering experiments which create zone-center phonons. This experiment is not easy, because the gap of the GaAs substrate is smaller than that of the GaAs/AlAs superlattices. We thus need to remove the substrate over a small area to define a window in the sample. The two studied samples, grown by molecular beam epitaxy at low temperature,[8] have the following parameters:

S1: $d=4.5$ nm, $x=0.56$,

S2: $d=3.7$ nm, $x=0.77$.

The chosen compositions correspond to the minimum (S1) and maximum (S2) predicted zone-center gaps. We chose small-period samples in order to increase both the folded average frequency and the direct band gap. This indeed allows us to work in the transparency range of the superlattice just below the direct absorption edge in order to maximize the Raman signal. These conditions are obtained for an incident wavelength of 676 nm and a sample temperature of 125 K for S1 and, respectively, 567 nm and 250 K for S2 (krypton-ion laser lines).

In these conditions, the usual backscattering result (a doublet) is obtained when focused out of the window area. In the same backscattering configuration, but on the window area, a new line appears in the S1 spectrum, located, within experimental accuracy, at the doublet average frequency [see Fig. 2(a), lower curve]. We attribute this additional line to forward scattering from the zone-center phonon in this sample with vanishing acoustical gap. The onset of forward-scattering lines in the backscattering configuration is due to reflections from the exit surface of the layer. In the forward-scattering experimental configuration [see Fig. 2(a), upper curve], the relative intensity of the lines is modified, the backscattering contribution coming now from an internal reflection. The results obtained on S2 are shown in Fig. 2(b). The backscattering spectrum (lower curve) contains two lines as usual; the forward-scattering spectrum (upper curve) only one which lies now between the doublet average frequency and the backscattering higher-component frequency. The contribution of the reflected beams is not so clear for this sample: They appear only as a low-energy (high-energy) shoulder in the backscattered (forward-scattered) spectrum. All the lines on both samples have been recorded in $z(x,x)z$ configuration. We never get any

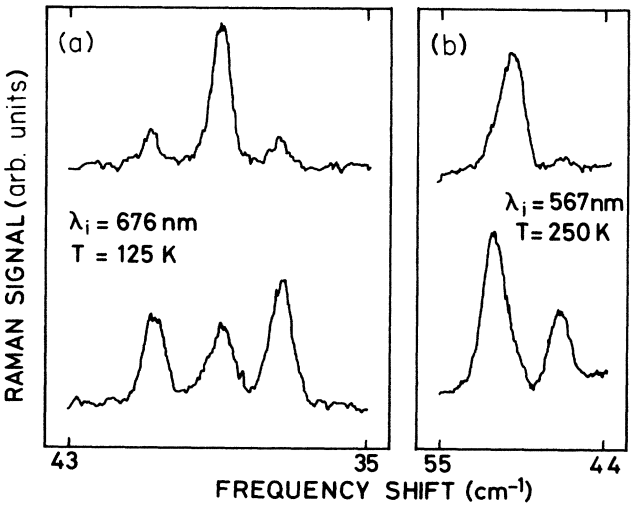

FIG. 2. Raman spectra on samples (a) S1 and (b) S2. In both cases the upper (lower) curve corresponds to the forward (backward) configuration.

signal in the $z(x,y)z$ one.

In order to understand these results, we must analyze the symmetry and scattering intensity of the folded lines. Each zone-center doublet is made of a symmetric and an antisymmetric mode: The displacement field related to the symmetric (antisymmetric) mode is symmetric (antisymmetric) relative to each midlayer plane. The upper energy component of the first zone-center doublet corresponds to the symmetric mode if $x < x_c$ and to the antisymmetric one if $x > x_c$. The symmetry of the photoelastic tensor in cubic crystals forbids the observation of any line in the $z(x,y)z$ configuration. As a consequence, the intensity scattered from the superlattice, considered as the sum of contributions of each layer (displaying local cubic symmetry), also vanishes in the $z(x,y)z$ configuration. Moreover, the scattering activity of the symmetric modes vanishes also in $z(xx)z$ configuration because of the symmetric character of the modulated photoelastic tensor relative to the midlayer planes. Furthermore, as one moves away from the Brillouin-zone center, the two modes mix together, and the scattering activities in parallel configuration of the two components of the doublet progressively become equal. At a given wave vector, the mixing increases as the zone-center gap decreases.[7]

This analysis agrees with all present and published[1–4,8] results. In backscattering experiments the phonon wave vector is always of the order of 10^6 cm^{-1}, a value which usually corresponds to a complete mixing and thus to an equal scattering intensity for both doublet components. The only exception is GaAs/AlAs superlattices with very short period and values of x such that the gap contribution to the doublet splitting becomes comparable to the dispersion one. In samples with smaller values of x, the lower energy line dominates the spectrum,[1,8] whereas in S2 the upper one does. In this sample the vanishing intensity of the lower energy line in forward scattering agrees with the strict zone-center selection rules. From the position of both the backscattering lines and the forward-scattering one, we can deduce the value of the zone-center acoustical gap in this sample, a value which fits very well the model calculation, as can be seen in Fig. 1 where all our experimental results are shown. An overall agreement is obtained.

In conclusion, we have reported the first evidence by Raman forward scattering of a folded acoustical gap at zone center. The values of the gap opening are in rather good agreement with the elastic-model predictions. The observed selection rules well support the assignment of the photoelastic mechanism as the dominant scattering process. A more detailed study of this point is in progress.

It is a pleasure for us to acknowledge G. Le Roux for careful x-ray diffraction measurements and A. Constant, R. Bertrand, and J. Charil for technical assistance. The Centre National d'Etudes des Télécommunications is "Unité associée au Centre National de la Recherche Scientifique (UA250)."

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