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Experimental evidence for coupled-mode phonon gaps in superlattice structures

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Frequency gaps for acoustic phonons with a propagation direction oblique to the superlattice layers are investigated by phonon transmission spectroscopy in amorphous superlattices. In addition to the gaps at the center and at the boundaries of the superlattice mini-Brillouin-zone, the phonon transmission spectra show gaps associated with stop bands for coupled phonon modes with mixed longitudinal and transverse character. The position of the gaps is well explained by a description of the phonons in the elastic continuum limit.

The periodic alternation of layers with different acoustic properties in a superlattice has important consequences for the propagation of acoustic phonons.[1] The most important of these is the folding of the phonon dispersion relation along the direction perpendicular to the layers (z direction) into a mini-Brillouin-zone of dimension $2\pi/d$, where d is the repetition period.[2] The zone-folding effect is a consequence of the artificial periodicity imposed by the growth process. It has been experimentally verified in the case of phonons propagating along the z direction through the observation of the so-called folded-phonon doublets in the low-frequency Raman spectrum of crystalline[2] and amorphous[3] superlattices.

A second consequence of the artificial periodicity is the opening of frequency gaps in the phonon dispersion relation.[4-7] The gaps result from the lifting of the degeneracy of phonon modes at the points where different branches of the zone-folded phonon dispersion cross. Modes from the different branches interact provided they have the same symmetry and the solution of the wave equation for frequencies within the gap corresponds to nonpropagating modes with a complex wave vector. Acoustic vibrations propagating perpendicular to the superlattice layers are classified into one longitudinal (LA) and two orthogonal transverse (TA) modes; the latter two are degenerate for isotropic layers. In this case, frequency gaps open up at the center and at the boundary of the mini-Brillouin-zone where the branches of either the longitudinal or the transverse modes touch. The existence of these gaps has been experimentally demonstrated by phonon transmission[4,6,7] and Raman scattering spectroscopy.[7,8]

In this Rapid Communication we present experimental evidence from phonon transmission spectroscopy measurements for additional frequency gaps in the spectrum of acoustic phonons in a-Si:H/a-SiN$_x$:H superlattices. These gaps are associated with phonon modes with a wave-vector component in the direction parallel to the layers (x direction). The phonon eigenmodes in these propagation directions are the following: one pure transverse shear wave polarized along the y direction and two coupled modes with mixed LA and TA character polarized in the (x,z) plane. The mode mixing is a consequence of the nonzero q_x component of the phonon wave vector.

We show in Fig. 1 the dispersion relation of the coupled LA-TA modes in an a-Si:H/a-SiN$_x$:H superlattice calculated using the continuum elastic model presented in Ref. 9. In this model each amorphous layer of thickness t and density ρ is assumed to be isotropic and characterized by a longitudinal sound velocity v^{LA} and a transverse sound velocity v^{TA}. The sound velocities used in the calculation were extracted from Ref. 7 and the results shown in Fig. 1 are for a superlattice with 8.5-nm period and with a thickness ratio of the silicon to silicon-nitride layers equal to 1.5. For a vanishing wave-vector component in the direction parallel to the layers ($q_x=0$) the modes are uncoupled and the dispersion is simply the superposition of the pure LA and TA dispersions. They exhibit frequency gaps only at the center $q_z=0$ and at the boundary $q_z=\pi/d$ of the superlattice mini-Brillouin-zone. Away from these gaps, the dispersion relations are linear and the LA and TA branches cross at the frequencies ω_l^x given by

$$\omega_l^x=\frac{2}{1\pm r_v}\bar{\omega}_l^{\mathrm{TA}} . \tag{1}$$

Here r_v is the ratio of the effective sound velocities for transverse and longitudinal phonons propagating along the z direction, and $\bar{\omega}_l^{\mathrm{TA}}$ is the center frequency of the lth gap of the pure TA dispersion. For a nonvanishing q_x component the waves have mixed LA-TA character and they can therefore interact: Modes from adjacent branches repel each other when their frequencies approach the crossing frequency of the two folded branches. Thus additional gaps corresponding to an imaginary q_z component open up in the (ω,q_z) dispersion that we shall call internal gaps to distinguish them from the main gaps at the center and at the boundary of the mini-Brillouin-zone. Note that for a small q_x component the internal gaps are centered at the frequencies ω_l^x given by Eq. (1). These additional gaps have also been calculated recently for crystalline GaAs/AlAs superlattices by Tamura and Wolfe.[10]

Our measurements were performed on glow-discharge

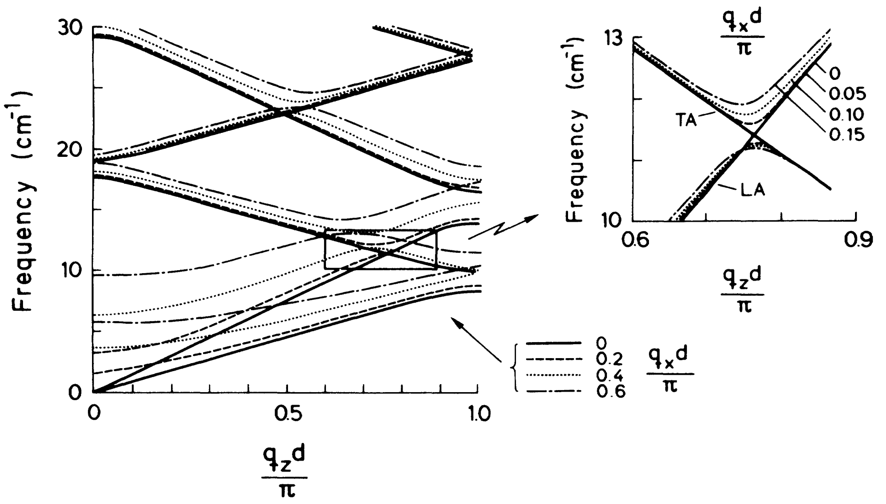

FIG. 1. Dispersion relation projected in the (ω, q_z) plane for the coupled modes with mixed LA and TA character and wave vector $(q_x, 0, q_z)$ in an a-Si:H/a-SiN$_x$:H superlattice. The parameters for silicon hydride (silicon nitride) used in the calculation were the following: Thickness 5.0 nm (3.5 nm), density 2.32 g/cm^3 (2.32 g/cm^3), LA sound velocity 7.1×10^5 cm/s (9.6×10^5 cm/s), and TA sound velocity 4.3×10^5 cm/s (5.8×10^5 cm/s).

a-Si:H/a-SiN$_x$:H superlattices prepared as described elsewhere.[3] For the phonon transmission measurements the superlattices where grown on the (111) surface of 4-mm-thick crystalline silicon substrates. The phonons were generated by a 1×1-mm^2 Al tunnel junction deposited on the back side of the substrate and detected by a 1×1-mm^2 Sn tunnel junction on top of the superlattice as indicated in the inset of Fig. 2. Using special modulation techniques this configuration allows the spectral measurement of the phonon transmission above 9.5 cm^{-1}.

Figure 2(a) shows the phonon transmission spectrum for a superlattice consisting of 130 repetition periods of 8.5 ± 0.5 nm each. The calculated dispersion for the coupled LA-TA modes of this sample is shown in Fig. 1. The signal above 9.5 cm^{-1}—the phonon detection threshold of the Sn tunnel junction—corresponds to acoustic-phonon transmission through the sample. The transmission minima in this region are attributed to frequency gaps in the zone-folded dispersion relation for acoustic phonons.[4] This conclusion is based on the observation that the positions of these minima scale with the superlattice period.[7] Incoming phonons with frequencies within the gaps are constructively Bragg reflected at the superlattice interfaces and therefore filtered out of the transmission spectrum.

In order to assign the transmission minima to the frequency gaps in the dispersion relation we also measured the low-frequency Raman spectrum of the same sample in backscattering configuration, as shown in Fig. 2(b). The sharp peaks in this spectrum correspond to the excitation of the two lowest LA folded branches at $q_z=6.2\times10^5$ cm^{-1} for modes propagating perpendicular to the layers.[2,3] The TA modes are not observable in this geometry.[5] The average frequency of the doublet corresponds to the center frequency $\bar{\omega}_2^{\mathrm{LA}}$ of the second LA gap (i.e., the lowest zone-center gap). There is no structure in the phonon transmission spectrum corresponding to this gap. This is probably due to the low sensitivity of the phonon transmission measurements for LA vibrations, as will be discussed below. The lowest-frequency LA gap at the boundary of the mini-Brillouin-zone is located at one half of the average frequency of the Raman doublet. This gap may, in principle, account for the minimum at 15.8 cm^{-1} in the phonon transmission spectrum. However, in the same frequency range one also expects a replica of the structure at 19.4 cm^{-1} due to the thermal current in the generator junction.[11]

The most prominent minimum at 19.4 cm^{-1} and the structure at one half of this frequency, identified by the concave shape of the absorption threshold of the Sn junction, are assigned to the frequency gaps for TA phonons propagating perpendicular to the layers.[7] The center frequencies of the LA and TA gaps discussed above are proportional to the corresponding superlattice effective sound velocities along the direction perpendicular to the layers. By comparing the Raman and the phonon transmission spectrum we calculate a ratio $r_v=v^{\mathrm{TA}}/v^{\mathrm{LA}}$ equal to 0.62 ± 0.03. This agrees with the ratio found in most semiconductors.

Transmission minima corresponding to the higher-frequency gaps are not observed in the phonon transmission spectrum. This might be due to strong phonon scattering in this high-frequency range which destroys the phase coherence necessary for constructive reflection at the interfaces. The higher generation rate of TA phonons in the Al junction in comparison with LA modes accounts for the larger reduction in phonon transmission within the TA gaps.[11] In addition, the transverse T2 phonons are strongly focused on the detector owing to the anisotropic propagation on the (111)-oriented silicon substrate, whereas the longitudinal modes are only weakly focused.[12]

The transmission minima discussed above correspond to the type of frequency gaps previously observed in phonon transmission spectra.[4,6,7] The spectrum of Fig. 2(a) exhibits, however, two additional minima: a sharp and nar-

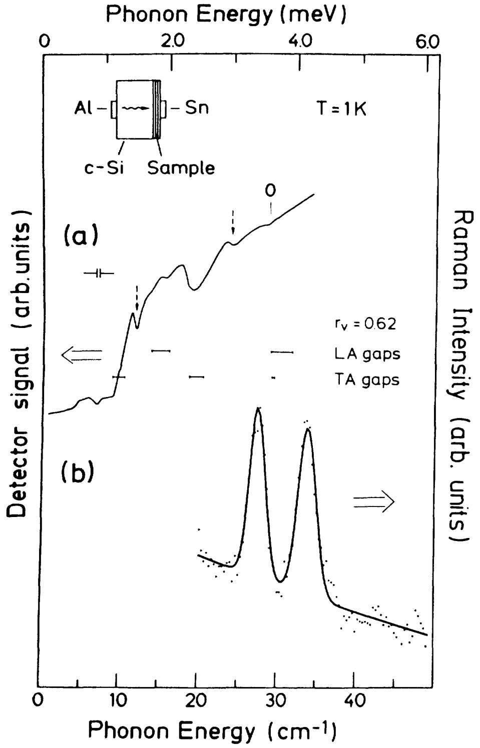

FIG. 2. (a) Phonon transmission and (b) Raman spectrum of an a-Si:H/a-SiN$_x$:H superlattice with a repetition period of 8.5 ± 0.5 nm. The bars below the phonon transmission spectrum indicate the position of the frequency gaps for TA and LA phonons. The arrows indicate the gaps for coupled modes with mixed LA-TA character. The structure at 29.3 cm^{-1} in the phonon transmission spectrum is associated with the vibration of interstitial oxygen in the crystalline silicon substrate (Ref. 11).

row one at 12.1 cm^{-1} and a much broader feature at 24.2 cm^{-1}. We attribute these minima to the lowest internal gaps for coupled LA-TA modes with propagation direction deviating from the superlattice normal as discussed in connection with Fig. 1. The phonons acquire the nonzero wave-vector component through oblique incidence at the substrate-superlattice interface or through elastic scattering processes within the superlattice. The incidence angle at the interface is smaller than 20° as determined from the geometric arrangements of the generator and detector junctions. This corresponds to a maximum q_x of $\sim 0.2(\pi/d)$ for a q_z about halfway between the center and the edge of the mini-Brillouin-zone. Note that even for such small values of q_x the width of the internal gaps can be relatively large (1 cm^{-1}), as deduced from the inset of Fig. 1.

The positions of the additional transmission minima in Fig. 2(a) agree well with the frequencies in the dispersion relation of Fig. 1, where the branches of the coupled modes cross. The sharpness of the internal minimum at 12.1 cm^{-1} in Fig. 2(a) indicates that the x component of the phonon wave vector is restricted to a small range of values centered around $q_x = 0$. The inset of Fig. 1 shows that in this case the internal gaps are centered at the cross frequencies ω_l^x of the pure LA and TA dispersions given by Eq. (1). In Fig. 3 we plot for a series of superlattices with different periods the observed center frequency of the first ($l=1$) and of the second ($l=2$) internal gaps, $\bar{\omega}_l^{\text{LA-TA}}$ as a function of the corresponding frequencies of the pure TA gaps, $\bar{\omega}_l^{\text{TA}}$. The experimental points lie on a straight line in concordance with the predictions of Eq. (1) for the lowest-frequency gaps [plus sign in Eq. (1)]. The slope of this line gives a value of 0.59 ± 0.01 for the ratio of the effective sound velocities for transverse and longitudinal vibrations propagating perpendicular to the layers. This result agrees well with the value $r_v = 0.62 \pm 0.03$ determined from the comparison of Raman and phonon transmission data.

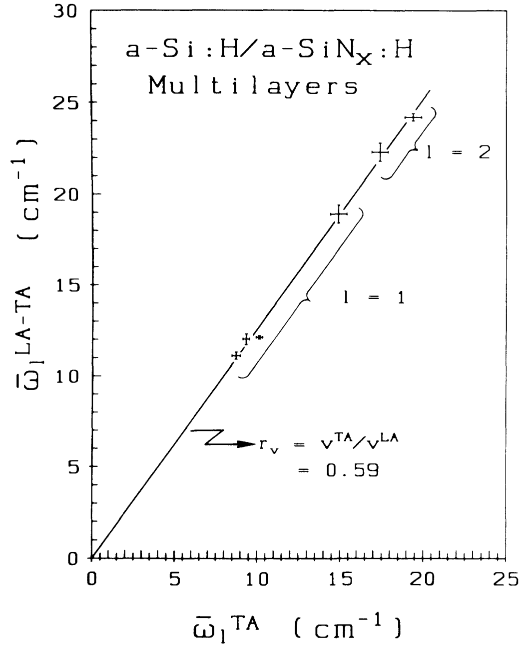

FIG. 3. The center frequency of the lth internal gap for coupled modes, $\bar{\omega}_l^{\text{LA-TA}}$ plotted vs the center frequency of the lth TA gap, $\bar{\omega}_l^{\text{TA}}$ in a-Si:H/a-SiN$_x$:H superlattices with different periods.

In conclusion, we report experimental evidence for new stop bands in the phonon transmission spectra of amorphous superlattices. These stop bands are associated with phonons with mixed LA and TA character that propagate in a direction deviating from the superlattice normal. Further investigations of the position and of the width of these gaps as a function of the orientation of the phonon beam are currently underway.

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[1] M. Cardona, in *Lectures on Surface Science,* edited by G. R. Castro and M. Cardona (Springer-Verlag, Heidelberg, 1987), p. 2; M. V. Klein, IEEE J. Quantum Electron. **QE-22**, 1760 (1986).

[2] C. Colvard, R. Merlin, M. V. Klein, and A. C. Gossard, Phys. Rev. Lett. **45**, 298 (1980).

[3] P. Santos, M. Hundhausen, and L. Ley, Phys. Rev. B **33**, 1516 (1986).

[4] V. Naranayamurti, H. L. Störmer, M. A. Chen, A. C. Gossard, and W. Weigmann, Phys. Rev. Lett. **43**, 2012 (1979).

[5] C. Colvard, T. A. Grant, M. V. Klein, R. Merlin, R. Fisher, H. Morkoc, and A. C. Gossard, Phys. Rev. B **31**, 2080 (1985).

[6] O. Koblinger, M. Mebert, E. Dittrich, S. Döttlinger, and W. Eisenmenger, in *Phonon Scattering in Condensed Matter,* edited by A. C. Anderson and J. P. Wolfe (Springer-Verlag, Berlin, 1986), p. 156.

[7] P. V. Santos, L. Ley, M. Mebert, and O. Koblinger, in *Interfaces, Superlattices, and Thin Films,* Materials Research Society, Vol. 77, edited by J. D. Dow and I. K. Schuller (Materials Research Society, Pittsburg, PA, 1987).

[8] B. Jusserand, F. Alexandre, J. Dubard, and D. Paquet, Phys. Rev. B **33**, 2897 (1986).

[9] R. Hotz and R. Siems, Solid State Commun. **51**, 793 (1984).

[10] S. Tamura and J. P. Wolfe, Phys. Rev. B **35**, 2528 (1987).

[11] W. Eisenmenger, in *Physical Acoustic,* edited by W. A. Mason and R. N. Thunston (Academic, New York, 1976), Vol. XII, p. 79.

[12] D. Marx and W. Eisenmenger, Z. Phys. B **48**, 277 (1982).