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Observation of folded-zone acoustical phonons by Raman scattering in amorphous Si-SiN_x superlattices

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The excitation of folded-zone longitudinal acoustic phonons in a-Si:H–a-SiN_x:H periodic multilayer structures is observed by Raman scattering. The energies of the two lowest folded phonon branches are followed by varying the lattice period and the laser frequency. The data are quantitatively explained by a description of the phonons in the elastic continuum limit.

Periodic multilayer structures consisting of alternating layers of amorphous hydrogenated silicon (a-Si:H) and amorphous hydrogenated silicon nitride (a-SiN_x:H) have been fabricated recently.[1-3] They are the amorphous analogs of the well-known crystalline superlattices[4] and combine thus in a unique way the homogeneity and isotropy of the amorphous constituent materials with the tailored one-dimensional periodicity imposed by the growth process. Previous investigations have concentrated on properties of the "amorphous superlattices" that do not reflect their periodicity but depend rather on the juxtaposition of narrow (a few tens of angstroms) slabs of material with different band gaps and the ensuing modifications of their electrical and optical properties.[1-3]

In this paper we present experimental evidence for the modification of the longitudinal acoustical (LA) phonon dispersion curves by the periodic modulation of the elasto-optic properties in a-Si:H–a-SiN_x:H superlattices. The folding of the LA phonon branch along the direction perpendicular to the layers into a Brillouin zone of dimension $2\pi/d$, where d is the period of the superlattice, gives rise to new peaks in the Raman spectra which are unobservable in unstructured material.[5] Gaps occur at the center and the boundaries of the new Brillouin zone as a result of the periodic modulation of the density and the elastic constant.[6,7] We show that the lowest gap at $q=0$ can be inferred from a systematic investigation of the LA phonon scattering as a function of the superlattice period d.

The samples considered here consist of a-Si:H–a-SiN_x:H superlattices grown by the glow-discharge decomposition of silane and ammonia in the same apparatus used for the preparation of doping-modulated multilayer structures.[8] They have periods ranging from 60 to 190 Å and a total thickness of the order of 1 μm. The composition modulation was achieved by alternating the gas composition in the discharge chamber from SiH_4, used to grow the a-Si:H layers, to a mixture of SiH_4 (1 part) and NH_3 (5 parts per volume) for the a-SiN_x:H layers. This composition leads to quasistoichiometric silicon nitride layers, with $x \simeq 1.3$. The gas changes were performed without extinguishing the plasma. The thickness of the individual layers was adjusted through the deposition times after appropriate calibration. For the samples with periods smaller than 100 Å the discharge chamber was purged with argon between each gas change to minimize cross contamination. Raman measurements were performed in the backscattering configuration, with an incident angle of 45°, which corresponds to an angle of 14° relative to the surface normal inside the sample. The scattered light emerging perpendicular to the sample surface was collected and analyzed in a Jarrell-Ash double monochromator and its intensity measured in the photon-counting mode. In samples with periods greater than 100 Å, the Raman peaks to be discussed lie closer than 30 cm^{-1} to the laser line, and a third monochromator was used to reduce the elastic background. The samples were kept in vacuum (below 10^{-5} Torr) and at room temperature during the measurements.

Figure 1 shows a series of Raman spectra for amorphous superlattices with periods d between 57 and 190 Å. We as-

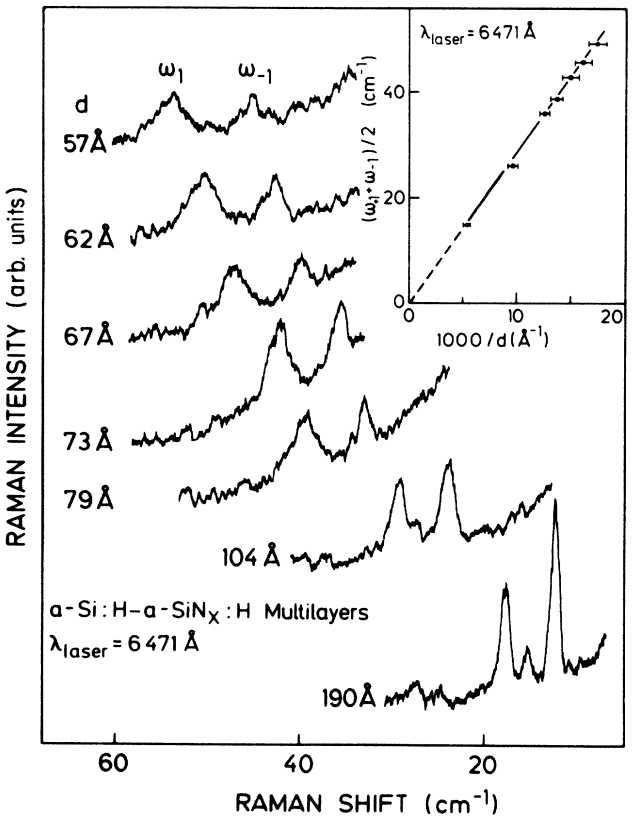

FIG. 1. Raman spectra for a-Si:H–a-SiN_x:H superlattices. The inset shows the dependence of the average frequency $(\omega_{+1}+\omega_{-1})/2$ on the inverse superlattice period $1/d$.

sign the two peaks which shift to lower energies with increasing superlattice period to the excitation of longitudinal acoustic phonons propagating in a direction perpendicular to the sample surface (z direction). The artificial periodicity in that direction gives rise to umklapp scattering processes with umklapp vectors of magnitude $m(2\pi/d)$ ($m=\pm1$, ±2, . . .). This yields the wave-vector conservation rule for the z component of the phonon wave vector Q:

$$Q = q + m\frac{2\pi}{d} \quad , \tag{1}$$

where $q = 2k_{\text{laser}}$ is the momentum transfer along z due to the backscattered light. We assign the observed doublets in Fig. 1 to excitations corresponding to $m=-1$ (ω_{-1}) and $m=+1$ (ω_{+1}), respectively. The excitation with $m=0$ is observed in conventional Brillouin scattering on unstructured a-Si:H.[10]

The dispersion relation of the longitudinal acoustic phonons in a superlattice

$$\omega(Q) = \omega(q + m2\pi/d) \equiv \omega_m(q); \tag{2}$$

$$m = 0, \pm1, \pm2, \ldots ,$$

exhibits, in general, gaps at the boundaries of the Brillouin zone [$q = m(\pi/d)$]. In the absence of gaps Eq. (2) represents simply the folding of the normal linear relationship between ω and Q with the effective sound velocity of the compound medium,[9] v_{eff}, as a proportionality constant:

$$\omega_m(q) = v_{\text{eff}}(q + m2\pi/d) \quad , \tag{3}$$

$$v_{\text{eff}}^2 = (c_1d_1 + c_2d_2)/(\rho_1d_1 + \rho_2d_2) \quad ,$$

where c_1, c_2, ρ_1, ρ_2, d_1, and d_2 are the elastic constants, densities, and thicknesses of the a-Si:H and a-SiN$_x$:H layers, respectively. For a fixed wave vector q the average frequency of branches -1 and 1 $(\omega_{-1}+\omega_{+1})/2$, should thus vary linearly with $1/d$. The linear relationship is maintained for a realistic dispersion relation as long as the ω_{+1} and ω_{-1} branches are disposed symmetrically with respect to (3). This expectation is borne out by the data as illustrated in the inset of Fig. 1. From a linear regression of the data points we obtain $v_{\text{eff}} = (8.5 \pm 0.5) \times 10^5$ cm/sec.

We followed the actual phonon dispersion in a particular sample as a function of q by varying the laser wavelength λ_{laser} and thereby $q = 4\pi n/\lambda_{\text{laser}}$, where n is the refractive index of the sample. The positions of ω_{-1} and ω_{+1} are plotted in Fig. 2 for a 100 Å a-Si:H–90 Å a-SiN$_x$:H sample as a a function of q. The data are quantitatively described by the layered continuum model for longitudinal elastic waves as developed by Rytov[6] and used by Colvard *et al.*[9] for the analysis of folded acoustic phonons in crystalline GaAs-AlAs superlattices. The quantities that enter in the model are $d_1/d_2 = 100/90$, the ratio of the thicknesses of the a-Si:H and a-SiN$_x$:H layers, ρ_1/ρ_2, the ratio of their densities, and v_1/v_2, the sound-velocity ratio. Using $\rho_1/\rho_2 = 0.68$ from the corresponding crystalline phases and $v_{\text{eff}} = 8.5 \times 10^5$ cm/sec, a good fit of the dispersion curve to the experimental data points is obtained for $v_1/v_2 = 0.83$. This corresponds to a sound velocity in the a-Si:H layers equal to 7.7×10^5 cm/sec, a value that is comparable to the sound velocities determined in unstructured a-Si:H by Brillouin scattering[10] (see Fig. 2).

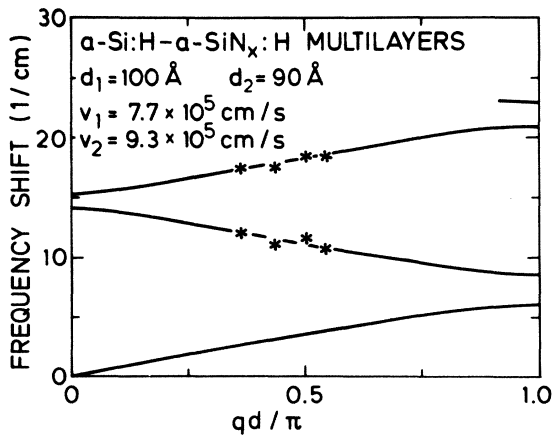

FIG. 2. Calculated dispersion of longitudinal acoustic phonons in a 100 Å a-Si:H–90 Å a-SiN$_x$:H multilayer. The data points were measured using different laser wavelengths.

The calculation gives a gap $\Delta\omega(0)$ of about 1 cm^{-1} at the center of the Brillouin zone. The zone-center gap scales with $1/d$ in this model for a given combination of materials and fixed d_1/d_2. Additional evidence for the existence of this gap is provided if we consider the line splitting

$$\Delta\omega(q) = \omega_{+1}(q) - \omega_{-1}(q) \quad , \tag{4}$$

as a function of $1/d$ for a fixed wave vector q.

In Fig. 3 $\Delta\omega$ is plotted versus $1/d$ for a number of samples with thickness ratios $d_1/(d_1+d_2)$ between 0.53 and 0.61. The increase in $\Delta\omega(q)$ with $1/d$ reflects the increasing influence of the zone-center gap as $qd \to 0$. The data points lie within the range calculated in the framework of the continuum model. In the absence of a zone-center gap $\omega(q)$ is given by Eq. (3) and $\Delta\omega(q) = 2v_{\text{eff}}q$ is independent of $1/d$.

In summary, we have demonstrated that folded-zone acoustic phonons can be observed in periodic multilayer structures based on amorphous semiconductors. We have shown that the dispersion of the folded-zone acoustic pho-

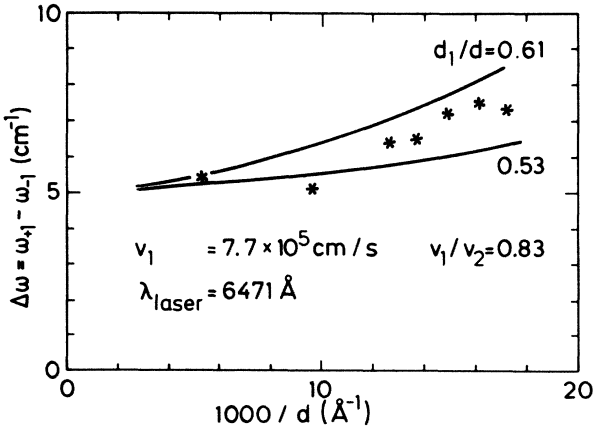

FIG. 3. The frequency difference $\Delta\omega(q) = \omega_{+1}(q) - \omega_{-1}(q)$ measured for fixed q as a function of the inverse superlattice period $1/d$. The solid lines are calculated in the continuum model for the two extreme ratios $d_1/(d_1+d_2)$ that bracket the corresponding values in the multilayers.

nons, and, in particular, the lowest zone-center gap, is well described by a continuum theory for the longitudinal acoustic phonons.

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