Raman scattering by folded *TA* and *LA* phonons in Si–$Si_{0.5}Ge_{0.5}$ superlattices

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The Raman scattering by acoustic phonons in Si–$Si_{0.5}Ge_{0.5}$ superlattices with axis along the (111) direction has been studied. The spectra reveal, in addition to the well-known lines of folded *LA* phonons, an allowed scattering by folded *TA* phonons. This is the first observation of the latter type of scattering.

The spectra of various superlattices are presently being studied by the Raman-scattering technique in a very active research effort.[1–5] It has been established that the existence of a superperiod results in a decrease in the size of the Brillouin zone along the axis of the superlattice. It has also been established that the phonon spectrum can be found in a first approximation by reducing (folding) the bulk spectrum into this new Brillouin zone of size π/D, where D is the period of the superlattice. As a result, additional wave branches arise in the frequency region of acoustic phonons. These so-called folded phonons are observed as equidistant doublets in scattering spectra. Various superlattices, e.g., GaAs–AlAs, Si–$Si_{1-x}Ge_x$ and Si–Ge superlattices, have been studied in Refs. 1–5 and some other papers of which we are aware. These superlattices were grown along the (100) crytallographic direction. In accordance with the form of the photoelastic-constant tensor (Ref. 6, for example), which determines the intensity of the scattering of light by acoustic phonons, research on superlattices in this orientation has resulted in the observation of, and detailed studies of, folded *LA* phonons, while the intensity of the scattering by *TA* phonons is approximately zero in this geometry.

In the present letter we report a study of Raman scattering in Si–$Si_{0.5}Ge_{0.5}$ superlattices grown along the (111) direction, for which scattering by both types of acoustic vibrations is allowed, in contrast with the (100) case.

The Si–$Si_{0.5}Ge_{0.5}$ superlattices were synthesized by molecular-beam epitaxy in an ultrahigh-vacuum apparatus[7] at a residual gas pressure below 10–8 Pa. As substrates we used silicon wafers in the (111) orientation. After a preliminary oxidation and chemical treatment by the method of Ref. 8, they were heated in a slow flow of silicon at a temperature ~850°C. As a result, an atomically clean surface with a 7×7 superstructure formed. The silicon flux was produced by sublimation from a silicon wafer heated by a current flow. The germanium was evaporated from a crucible source with a crucible consisting of pyrolytic boron nitride. Before the synthesis of the superlat-

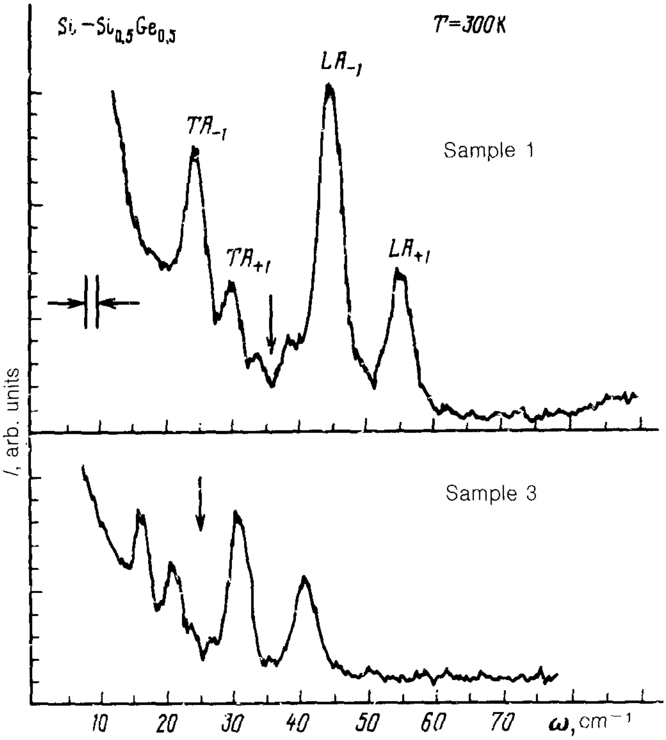

FIG. 1. Spectra of the Raman scattering by acoustic phonons of two Si–$Si_{0.5}Ge_{0.5}$ superlattices with periods $D_1^0 = 50$ Å and $D_2^0 = 72$ Å.

tices, we grew an epitaxial layer of silicon 1000 Å thick and then a buffer layer of a solid solution with a composition equal to the average composition over the thickness of the superlattice. The superlattices were grown at a substrate temperature of 500°C by periodically interrupting the germanium flux while holding the silicon flux constant. The thickness of the layers of silicon and the solid solution were monitored by measuring the oscillations in the intensity of the specular beam of electrons during reflection diffraction.[9] The composition was inferred from changes in the oscillation frequency upon the interruption of the germanium flux. The average growth rate of the superlattices was 100 Å/min; the number of layers was 10–30 atomic spacings; and the number of periods ranged up to 50.

The spectra reported in the present letter were excited by light with a wavelength $\lambda = 488$ nm and recorded by a DFS-52 spectrometer. During the measurements the samples were held in vacuum in order to eliminate parasitic lines in the low-frequency region due to scattering by rotational excitations of air (O_2 and N_2) molecules. Figure 1 shows scattering spectra in the frequency region of the acoustic vibrations of the two superlattices, for which the thickness of the Si layers, d_1, is equal to the thickness of the layers of the solid solution, d_2. For sample N_1 we had $D_1^0 = d_1 + d_2 = 50$ Å, and for the second sample we had $D_2^0 = 72$ Å. The overall thicknesses of the superlattices were 1500 Å and 3000 Å, respectively. In each spectrum (Fig. 1) we see two doublets, one of which (the lower-frequency one) is a consequence of scattering by folded *TA* phonons, while the other is a consequence of scattering by *LA* phonons. The distance between the lines in each doublet remains the same from sample to sample; in turn, the positions of the doublets depend on the period of the superlattice. The lines corresponding to scattering by bulk *TA* and *LA* phonons are not seen in our spectra because of the high level of background light near the laser line ($\omega = 0$).

To analyze the frequencies of the folded *TA* and *LA* phonons, we took the approach of Refs. 1–5, making use of the theoretical results of Rytov,[10] who derived an expression for the dispersion of acoustic waves in a medium in which the elastic properties vary periodically. The dispersion relations $\omega(q)$ are solutions of the equation

$$\cos(q, D) = \cos\left(\frac{\omega d_1}{v_1}\right)\cos\left(\frac{\omega d_2}{v_2}\right) - 1/2\,[R + 1/R]\sin\left(\frac{\omega d_1}{v_1}\right)\sin\left(\frac{\omega d_2}{v_2}\right) \qquad (1)$$

where $R = v_1\rho_1/v_2\rho_2$; ρ_1, ρ_2, v_1, and v_2 are the densities and sound velocities of the materials making up the superlattices; D is the period of a superlattice; and q is the wave vector along its axis. This equation is valid for phonons of both types (*LA* and *TA*). Since the elastic properties of Si and $Si_{0.5}Ge_{0.5}$ are approximately the same, we can use the substitution $1/2[R + 1/R] = 1 + \delta$ in (1), where $\delta \ll 1$. Setting $\delta = 0$, we then find the following solution of Eq. (1) for wave vectors q whose values are not too close to the center or boundary of the Brillouin zone:

$$\omega(q) = V_{SL}(2\pi/D)m \pm V_{SL}q, \qquad (2)$$

where $m = 0,1,2$, etc., is the index of the branch of folded phonons, and V_{SL} is the sound velocity in a superlattice, given by

$$V_{SL} = D[d_1^2/v_1^2 + d_2^2/v_2^2 + (R + 1/R)d_1 d_2/v_1 v_2]^{-1/2}\ . \tag{3}$$

The ± in (2) for a given m correspond to two different dispersion branches, which are observed as a doublet.

It follows from expression (2) that the distance between the lines of a doublet is given by $\Delta\omega(q_s) = 2V_{SL}q_S$, where $q_S = 2(2\pi n/\lambda)$ is the wave vector of the phonons which are participating in the scattering of the light (n is the refractive index). In our case, for $\lambda = 488$ nm, we have $q_S = 1.12\times10^6\ \mathrm{cm}^{-1}$. From the experimental values of the distances between the lines of the TA and LA doublets, $\Delta\omega^L = 10\ \mathrm{cm}^{-1}$ and $\Delta\omega^T = 5.4\ \mathrm{cm}^{-1}$, we find the sound velocities in Si–$\mathrm{Si_{0.5}Ge_{0.5}}$ superlattices to be $V_{SL}^L = 8.4\times10^5$ cm/s and $V_{SL}^T = 4.5\times10^5$ cm/s. In turn, the velocities of longitudinal and transverse sound waves in Si for the (111) direction are well known[11]: $V_1^L = 9.35\times10^5$ cm/s and $V_1^T = 5.05\times10^5$ cm/s. Using these values along with the values found for V_{SL}^L and V_{SL}^T, we find $V_2^T = (4.2\pm0.2)\times10^5$ cm/s and $V_2^L = (7.8\pm0.2)\times10^5$ cm/s for the $\mathrm{Si_{0.5}Ge_{0.5}}$ solid solution. These values were pre-

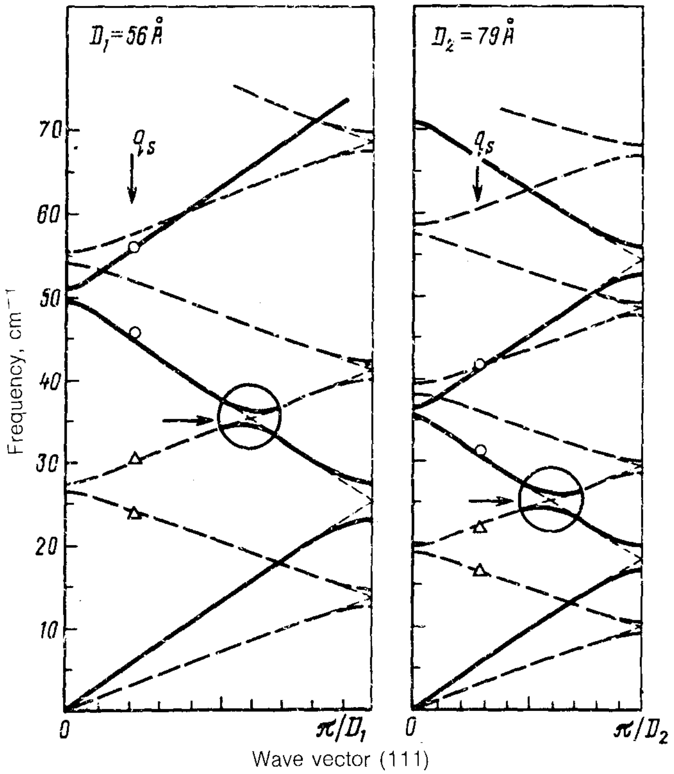

FIG. 2. Dispersion curves of folded LA phonons (solid lines) and folded TA phonons (dashed lines) calculated from expression (1) for the two superlattices. The experimental positions of the lines of the TA phonons are shown by triangles, those of LA phonons by circles. The arrows show the observed positions of the additional structural feature in the spectra.

viously unknown for the (111) propagation direction and have been determined here for the first time.

Figure 2 shows the dispersion $\omega(q)$ of folded *LA* phonons (solid lines) and *TA* phonons (dashed lines) according to calculations from (1) for two superlattice samples. In the calculations we varied the period D to fit the theoretical results to the experimental data. Also shown in this figure are the observed positions of the folded-phonon lines. The superlattice periods refined in this manner are $D_1 = 56$ Å and $D_2 = 78$ Å. They differe insignificantly from the values of D_1^0 and D_2^0 determined during the growth. It can be seen from Fig. 2 that the positions of the lines of folded *TA* and *LA* phonons are described well by expression (1). The calculated dispersion (Fig.2) deviates from linearity near the center and boundaries of the Brillouin zone. For phonons with $q = q_S$ these deviations are negligible. As a result, we can use the approximation which we used in determining the sound velocities in the superlattices from the distances between the lines of doublets.

In addition to the intense lines of folded phonons, our spectra reveal a faint structural feature, whose position is marked by the arrows in Fig. 1. It can be seen from Fig. 2 that in this frequency region (between the *TA* and *LA* doublets) the dispersion curves for the phonons cross. As a result of phonon-phonon coupling, the degeneracy is lifted, and the various phonon branches are renormalized, transforming into each other, as shown schematically in Fig. 2. A scattering by these phonons (with $q \neq q_S$) may be caused by a violation of wave-vector conservation. This explanation is supported by the shape of this structure in the spectra (Fig. 1): It has a characteristic dip, which corresponds to a zero density of phonon states in a gap which has formed. Furthermore, the position of the crossing point on these dispersion curves agrees well with experimental data.

In summary, the scattering of light in Si–Si$_{0.5}$Ge$_{0.5}$ superlattices with axis along the (111) direction makes it possible to observe both *LA* and *TA* folded phonons. The positions of the lines of the vibrations of both types can be described by the existing theory.[10] Furthermore, the spectra of these superlattices have structural features which may be associated with a coupling of *TA* and *LA* folded phonons.

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