Raman scattering by LA and TA phonons in GaAs/AlAs superlattices grown along the [111], [112], and [113] directions

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Raman scattering of light by longitudinal (LA) and transverse (TA) acoustic phonons has been studied in GaAs/AlAs superlattices grown on surfaces with indices other than (001). The frequencies and intensities of the Raman scattering induced by a modulation of the thickness of the layers in corrugated superlattices grown along the [113] direction have been determined.

It has recently become possible to grow high-quality GaAs/AlAs structures on surfaces with indices other than (001), specifically, (011) (Ref. 1), (012) (Ref. 2), (111) (Refs. 3 and 4), (112) (Ref. 5), and (113) (Refs. 6–8). Particularly interesting has been the discovery of a new effect:[7,8] an ordered microscopic faceting (corrugation) of heterojunctions in superlattices grown on surfaces with indices other than (001).

The first studies of Raman scattering by GaAs/AlAs superlattices (SLs) grown in the [011] (Ref. 1), [012] (Ref. 2), and [111] (Ref. 4) directions and corrugated SLs grown along the [113] direction[8] showed that the picture observed in these SLs is much more complex than that observed in SLs grown along the [001] direction. The reason lies in the low symmetry of these SLs and the appearance in the Raman scattering of oscillations which are forbidden in SLs on a (001) surface. The studies which we just cited focused on the frequency range of optical phonons. In the present letter we are reporting the first information on Raman scattering by acoustic LA and TA phonons in GaAs/AlAs SLs grown along the [111], [112], and [113] directions. Analysis of these results reveals the frequencies and intensities of the Raman scattering induced by a modulation of the thicknesses of the layers in corrugated SLs grown along the [113] direction.

The GaAs/AlAs SLs were grown by molecular beam epitaxy at a growth temperature of 660 °C. The growth was monitored with the help of reflection high-energy electron diffraction (RHEED). According to the RHEED data, the (111), (112), and (113) surfaces of GaAs and AlAs decompose into ordered arrays of microfacets at this temperature. For example, the (113) surfaces are ordered facets of microgrooves (corrugations) of height $\tilde{d}_z/2 = 10.2$ Å with a period $d_x = 32$ Å along the $[1\bar{1}0]$ direction. The SLs grown on a corrugated (113) surface are sets of thickness-modulated GaAs and AlAs layers.

The Raman spectra were excited by an argon laser ($\lambda_0 = 488$ nm; the power at the

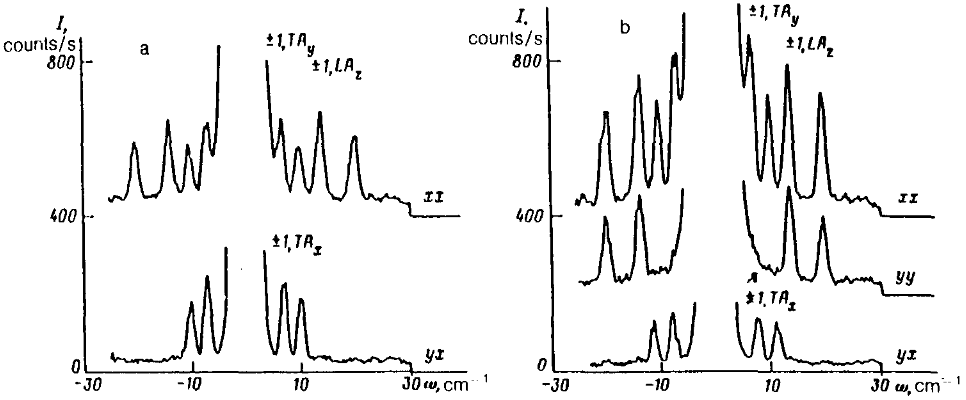

FIG. 1. Polarized Raman scattering spectra of GaAs/AlAs (55/55 Å) superlattices grown along (a) the [111] direction, and (b) the [112] direction.

sample was 50 mW in a spot $\approx 30\ \mu$m in diameter) and measured in a single-channel mode by a Raman spectrometer based on a Jobin–Yvon *U*-1000 double monochromator. The sample was in an atmosphere of gaseous helium at room temperature. Polarized Raman spectra were measured in a 180° geometry; the spectral width of the slit was 1.5 or 2 cm^{-1}.

A periodic modulation of the elastooptic constants in SL results in a scattering of light by acoustic phonons with wave vectors $\mathbf{Q}=m\mathbf{G}_z+\mathbf{q}$ (see the review by Jusserand and Cardona[9]). Here $m=\pm 1$ specifies the harmonic of the Fourier transform of the spatial distribution of the Ga (or Al atoms in the SL, $\mathbf{G}_z=\mathbf{z}2\pi/d_z$ is a reciprocal vector of the SL with a period d_z, and $\mathbf{q}=\mathbf{z}4\pi n/\lambda$ is the light scattering vector. We denote these phonons and the corresponding lines in the Raman scattering as m, LA_i and m, TA_i, where i specifies the displacement direction. In the coordinate system which we are using, the **z** axis is perpendicular to the plane of the SL, while the **x** axis is parallel to the [110] direction, which lies in the plane of all the SLs studied.

Figure 1 shows the spectra of Raman scattering from a GaAs/AlAs (55/55 Å) SL grown along the [111] direction. In the *xx* and *yy* polarizations here we see, in addition to the ± 1, LA_z doublet due to scattering by LA_z phonons with $\mathbf{Q}=\pm 1\mathbf{G}_z+\mathbf{q}$, a ± 1, TA_y doublet, which corresponds to TA_y phonons with the same wave vectors. In the *yx* polarization we see only the ± 1, TA_x doublet, which has the same frequencies and intensities as ± 1, TA_y.

Structures grown along the low-symmetry [112] and [113] directions have a latent optical anisotropy. This anisotropy can be seen well in the Raman spectra of a GaAs/AlAs (55/55 Å) SL grown along the [112] direction in Fig. 1b. The pattern observed in the *xx* polarization is similar to that observed in a superlattice grown along the [111] direction. In the *yy* polarization, on the other hand, the ± 1, TA_y lines are essentially absent, and the ± 1, LA_z lines are less intense by a factor of 1.5 than in the *xx*

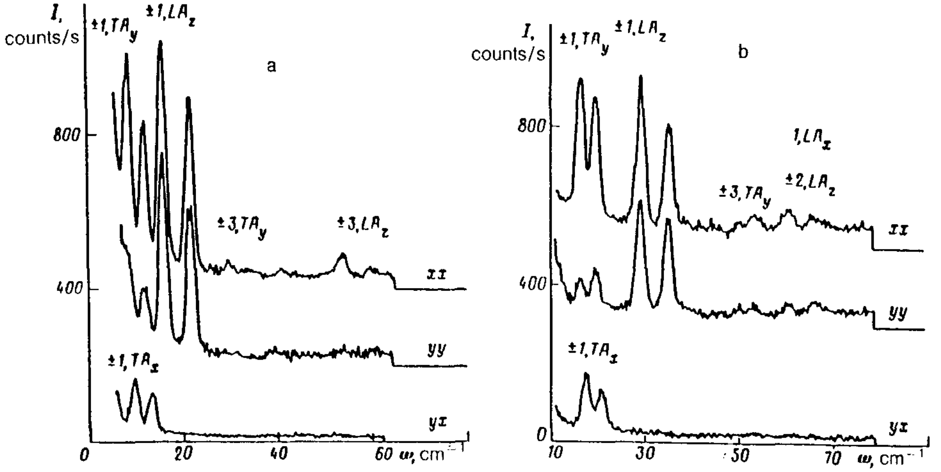

FIG. 2. Polarized Raman scattering spectra of GaAs/AlAs superlattices grown along the [113] direction. *a*—(47/47 Å); *b*—(27/27 Å).

polarization. In the *yx* polarization, we see only the ± 1, TA_x lines, whose frequencies are 10% higher than those of ± 1, TA_y. This difference stems from the lifting of the degeneracy from TA phonons propagating in a low-symmetry direction.

Figure 2 shows Raman spectra of two GaAs/AlAs SLs grown along the [113] direction. In this case we can also see a clear difference between the intensities of the Raman scattering in the *xx* and *yy* polarizations and a lifting of the degeneracy from the frequencies of the TA phonons. In addition to the intense ± 1, TA_y and ± 1, LA_z lines, in a (47/47 Å) SL we see some faint ± 3, TA_y and ± 3, LA_z lines (Fig. 2a). This is a scattering induced by the third harmonic of the SL potential. There are essentially no lines corresponding to the second harmonic in this SL, since they are forbidden in an SL with layers of equal thicknesses. In the (27/27 Å) SL, on the other hand, we see ± 2, LA_z lines (Fig. 2b), whose position allows us to assign them to the second harmonic. On the other hand, the more intense component of this doublet, denoted 1, LA_x in Fig. 2b, coincides in frequency with an LA_x phonon with a wave vector equal to the reciprocal vector of the corrugation of the (113) surface (more on this below). We accordingly assume that the scattering observed in this region is due to a modulation of the thickness of the layers in the SL.

The intensity of the Raman scattering by acoustic phonons in a SL is determined by the amplitudes of the Fourier transform of the spatial distribution of the elastooptic-constant tensor.[9] For the low-symmetry [211] and [311] directions, the LA_z and TA_y phonons are no longer purely longitudinal and purely transverse. The directions of the corresponding displacements and thus the intensities of the Raman scattering by these phonons also depend on the elastic-constant tensor of GaAs and AlAs.

We will not reproduce here the rather lengthy solution of this problem; we will simply discuss some results.

I. The frequencies of LA_z, TA_x, and TA_y phonons with $\mathbf{Q}=\pm\mathbf{G}_z+q$ calculated from the elastic-constant tensors of GaAs and AlAs (Ref. 10) agree well with the experimental data in all cases studied.

II. For superlattices grown along the [111], [112], and [113] directions, LA_z and TA_y phonons are allowed in the xx and yy polarizations, while TA_x phonons with $\mathbf{Q} \parallel \mathbf{z}$ are allowed in the yx polarization. The scattering intensity depends on three components of the tensor Δp_{ij} (this is the difference between the elastooptic constants of GaAs and AlAs). In the case $\mathbf{Q}\parallel \mathbf{z}$ the constants Δp_{11} and Δp_{44} appear in the linear combination $\Delta(p_{11}-2p_{44})$ in the expressions for the intensity. Analysis of the experimental results shows that the relative intensities of the Raman scattering depend only weakly on the quantity $\eta=\Delta(p_{11}-2p_{44})/\Delta p_{12}$, which we estimate to be $\eta=-0.1 \pm 0.1$.

III. A modulation of the thickness of the layers in SLs grown along the [113] direction leads to a scattering of light by LA_x, TA_y, and TA_z phonons (in this case we have $\mathbf{y}' \parallel$ [001], and $\mathbf{z}' \parallel$ [110]) with wave vectors $\mathbf{Q}_x \approx \mathbf{x}2\pi/d_x$, where $d_x=32$ Å is the period of the modulation of the layer thickness. The frequencies of these phonons are ≈ 59, 37, and 27 cm^{-1}, respectively.

IV. A scattering by an LA_x phonon is allowed in the xx and yy polarizations, while a scattering by TA'_y and TA'_z phonons is allowed in the yx polarization. The intensity of this scattering reaches a maximum when the period of the SL along the growth direction (d_z) is equal to the depth of the layer thickness modulation ($\tilde{d}_z$). A SL of this sort is a set of rhombic columns of GaAs and AlAs arranged in a checkerboard pattern and directed along the $\mathbf{y} \parallel [\bar{3}\bar{3}2]$ direction. In its Fourier transform, the amplitudes of the harmonics with $\mathbf{Q}_x=\mathbf{x}2\pi/d_x$ and $\mathbf{Q}_z=z2\pi/d_z$ are identical. The intensity ratio of the 1, LA_x, and 1, LA_z lines is thus determined by the elastooptic constants; it is $\approx[(4\rho+\eta+1)/2]^2/[(\eta+7)^2/66]$ in the xx polarization and $\approx[(9\eta+13)/22]^2/[(\eta+3.2)^2/18]$ in the yy polarization. Here $\rho=\Delta p_{44}/\Delta p_{12}$. With an increase in d_z, the intensity of the scattering by the 1, LA_z phonons increases, while that by the 1, LA_x phonons decreases, in proportion to $(\tilde{d}/d_z)^2$. In the GaAs/AlAs [113] (27/27 Å) SL which we studied, the intensity of the LA_x line in the yy polarization should be $\approx 4\%$ of the intensity of the ± 1, LA_z lines. If we assume $\rho \approx 0.5$, the calculated intensity of the LA_x line in the xx polarization is $\approx 12\%$ of ± 1, LA_z, in agreement with the experimental data.

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