Convolution of acoustic-phonon branches in GaAs/InAs superlattices

A. P. Shebanin, V. A. Gaĭsler, T. V. Kurochkina, N. T. Moshegov,
S. I. Stenin, and A. I. Toropov
Institute of Semiconductor Physics, Siberian Branch of the Academy of Sciences of the USSR

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A study of Raman scattering in GaAs/InAs superlattices, in which the ratio of the thicknesses of the GaAs and InAs layers is large, has made it possible to observe, for the first time, a convolution of the branches of acoustic phonons over a large part of the acoustic range. The dispersion for acoustic phonons in GaAs has been determined experimentally.

The existence of an additional periodicity in semiconductor superlattices causes marked changes in the photon spectrum of a crystal.[1] In particular, acoustic-phonon branches undergo a convolution within Brillouin minizones in the low-frequency part of the spectrum; the effect is manifested by several doublet peaks in the Raman spectra. According to Ref. 1, the intensities of these peaks are described by

$$I_{m_-^+} = A m^{-2} \sin^2 (m\pi d_1/d)[1 + n(\omega_{m_-^+})]\omega_{m_-^+}, \tag{1}$$

where m is the index of the doublet, ω is its frequency, the superscripts $+$ and $-$ on the number m specify the high-frequency and low-frequency peaks of the doublet, d is the period of the superlattice, d_1 and d_2 are the thicknesses of the neighboring layers ($d_1 + d_2 = d$), $n(\omega_m)$ is a Bose-Einstein distribution, and A is a proportionality factor. From relation (1) we can determine the decay of the intensity of the convolution of acoustic-phonon branches as m increases. In earlier studies, experiments were carried out on superlattices with $d_1 \approx d_2$. In this case the decay is pronounced according to (1): The intensity of the second doublet (and that of all even-numbered doublets) is zero. Because of the factor m^{-2}, the intensity of the third doublet is nearly an order of magnitude lower than that of the first; and so forth. In this letter we take up for the first time the limiting case of a superlattice in which the ratio of the layer thicknesses d_1 and d_2 is large. In such superlattices, at $kT > \hbar\omega$, and for values of m which are not very large, expression (1) can be put in the form

$$I_{m_-^+} = A\pi^2 d_1^2 d^{-2} kT\hbar^{-1}; \tag{2}$$

i.e., the intensity does not depend on the index m. It thus becomes possible to observe a convolution of acoustic-phonon branches over a significantly broader range. We have indeed observed these processes experimentally in the present study.

We studied $(\mathrm{GaAs})_k(\mathrm{InAs})_l$ superlattices grown by molecular epitaxy in apparatus developed in the Institute of Semiconductor Physics, Siberian Branch of the Academy of Sciences of the USSR. Here k and l are the numbers of monomolecular

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layers; $l = 2$ for all samples; and $k = 6$, 10, 19, and 22 for samples A, B, C, and D, respectively. The growth of each layer was monitored by high-energy electron diffraction and was terminated when the intensity of the specular reflection in the diffraction reached a maximum, which corresponded to a maximum filling of the upper monolayer.[2] The Raman scattering was studied on a U-1000 spectrometer with excitation by the light from an argon laser with a wavelength $\lambda_i = 514.5$ nm. The measurements were taken at $T = 295$ K in a vacuum chamber in the Brewster geometry of quasiback scattering.

Figure 1 shows Raman spectra of four samples. The dashed line shows some broad structural features which arise from the disordering of the crystal structure[1] (DATA and DALA).[1)] Against the background of these broad features we can clearly see doublets corresponding to a convolution of acoustic-phonon branches. The number

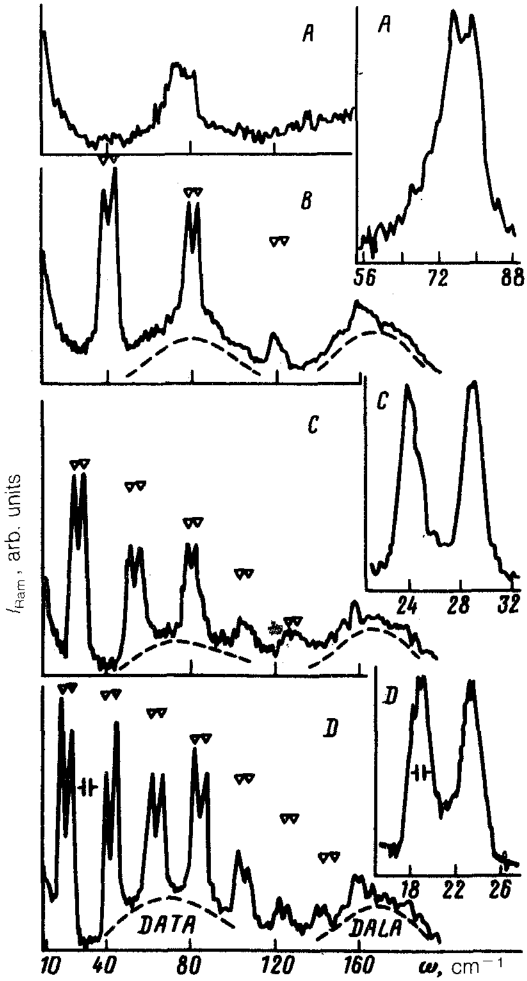

FIG. 1. Raman scattering spectra of GaAs/InAs superlattices with several periods. Sample A—$d = 23.0$ Å; sample B—$d = 34.3$ Å; sample C—$d = 59.7$ Å; sample D—$d = 68.2$ Å.

of these doublets increases with increasing $d_1 (d_2 = \text{const})$. In the spectrum of sample D $(d_1/d_2 = 11)$ we see seven of these doublets. The theoretical intensities and frequencies of these doublets are shown by the triangles in Fig. 1. In the low-frequency part of the spectra the theoretical intensities agree well with the experimental data. As we move into the frequency interval with the DALA structural feature, we see a noticeable discrepancy. The frequencies of these convolution peaks were calculated from an expression which we derived especially for superlattices in which d_2 is equal to two InAs monomolecular layers, which we take to be a liner chain of atoms. The d_1 layer, which is much thicker, is treated in the continuum approximation. The expression specifying the dispersion is

$$\cos(qd) = \cos(kd_1)\left[\frac{\psi-\alpha^2+\psi^2}{\alpha}\right] + \sin(kd_1)\left[\frac{(1+\psi)^2-\alpha^2+\alpha^2\delta^2-\psi^2\delta^2}{2\alpha\delta}\right], \qquad (3)$$

where q is the wave vector of a phonon in the superlattice, $K = \omega/v_1$, ω is the frequency of a phonon in the superlattice, v_1 is the velocity of an LA phonon in GaAs, $\alpha = (2 - M_{\rm As}\omega^2\beta^{-1})^{-1}$, $M_{\rm As}$ is the mass of the As atom, $\beta = 2C'_{11}a_0$ is the stiffness constant of the In-As atomic bond, C'_{11} is an elastic constant, a_0 is the lattice constant of InAs, $\psi = (M_{\rm In}\omega^2\beta^{-1} - 2 + \alpha)$, $M_{\rm In}$ is the mass of the In atom, $\delta = (KC_{11}a^2\beta^{-1})/2$, C_{11} is an elastic constant and a is the lattice constant of GaAs.

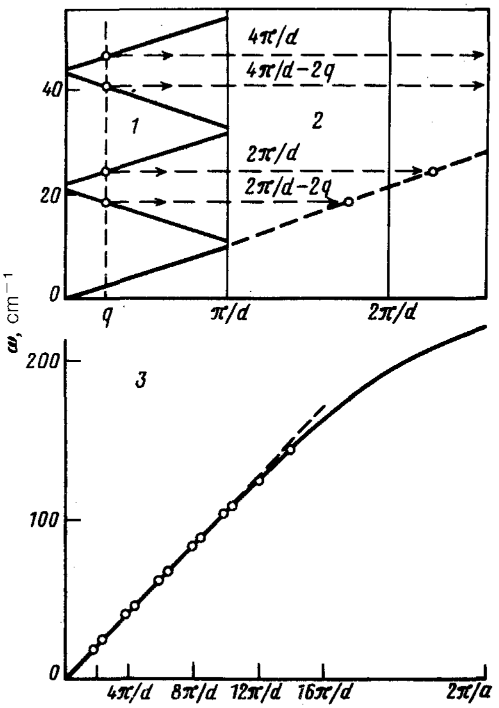

FIG. 2. Dispersion laws for phonons in GaAs/InAs superlattices and for LA phonons in GaAs.

We constructed dispersion laws $\omega(q)$ on the basis of (3) for all of the structures which we studied. These laws give a highly accurate description of the experimental data in terms of the values of the frequencies[2)] $\omega_{m\pm}$. The first sector in Fig. 2 shows the dispersion law and experimental values of the frequencies of the first two doublets of sample *D*. Carrying out an inverse convolution, and making use of the experimental values of $\omega_{m\pm}$ (the arrows in sector 2 in Fig. 2), we can construct a dispersion law for the *LA* phonons of the bulk material (sector 3 in Fig. 2). The solid line in sector 3 in Fig. 2 is the theoretical dispersion law for *LA* phonons of GaAs (Ref. 4).

Analysis of the experimental results shows that a slight change in the period of the supperlattice leads to a significant frequency shift of the convolution doublets (*C* and *D* in Fig. 1). It would thus be possible to work from Raman-scattering data to determine the diameter *d* of superlattices of interest within ~0.5 Å. Fluctuations Δd in the superlattice period lead to a nonuniform broadening of the convolution peaks, so the half-widths of the peaks can be used to estimate the diameter Δd. The insets in Fig. 1 show the first convolution doublets, which were recorded at the best resolution. The value found for Δd from the half-width of the convolution peaks does not exceed 1 monolayer.

In summary, a study of Raman scattering in GaAs/InAs superlattices with $d_1 \gg d_2$ has made it possible to observe a convolution of branches of acoustic phonons over a large part of the acoustic range and to construct a dispersion law for *LA* phonons in GaAs. Raman-scattering data can be used to determine the parameters *d* and Δd highly accurately.

[1)] DATA and DALA stand for "disorder-activated *TA* and *LA* phonons."

[2)] The dispersion laws found in accordance with Refs. 1 and 3 differ negligibly from those which we found. The differences for the first doublets are no greater than 0.3 cm^{-1}.

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