Confined optical phonons in the AlAs layers of GaAs/AlAs superlattices

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The phonon spectrum of high-quality GaAs/AlAs superlattices has been studied in the region of AlAs frequencies by a Raman-scattering method. Peaks corresponding to longitudinal optical phonons with a quantization order of 1–5 were resolved in the spectra. The experimental results support the results of a microscopic calculation of the dispersion of LO phonons of bulk AlAs.

Confined optical phonons in semiconductor superlattices are manifested in Raman scattering spectra as a series of peaks with frequencies corresponding to the frequencies of optical phonons of the bulk material, with wave vectors

$$q=\frac{m\pi}{(n+\delta)a_0},\quad m=1,2,3...,\tag{1}$$

where a_0 is the thickness of a monolayer, n is the number of monolayers of the corresponding material, and the parameter δ describes the penetration of a phonon into neighboring layers of the other material.[1,2] Raman-scattering data on the frequencies of confined optical phonons can thus be used to determine the dispersion of the optical phonons of the bulk forms of the materials forming the superlattice. This is an extremely important opportunity in the case of AlAs, since the instability of this material makes it difficult to study by the method of cold-neutron scattering.

Many studies of optical phonons confined in GaAs layers have been published.[1,2] The experimental values of the frequencies of these phonons agree well with the existing theoretical dispersion relations for bulk GaAs (Ref. 3). Pusep *et al.*[4] have reported the observation of confined TO phonons of AlAs in GaAs/AlAs superlattices by IR spectroscopy. Again, the frequencies of these phonons agree well with the theoretical dispersion of bulk AlAs (Ref. 3). However, the experimental data on LO phonons confined in AlAs layers[5–7] are very scanty and contradictory. Our purposes in the present study were to take a detailed look at the effects of the confinement of LO phonons in the AlAs layers by a Raman-scattering method and to analyze the dispersion of LO phonons in AlAs.

Figure 1a shows a typical Raman-scattering spectrum of GaAs/AlAs superlattices in the AlAs frequency region. This spectrum has peaks of LO and TO phonons, along with a broad spectral feature due to interface phonons. In the Raman-scattering spectra of an ideal superlattice grown in the (001) direction, only peaks of LO phonons should be seen. In real superlattices, with rough interfaces, interface phonons are manifested in the spec-

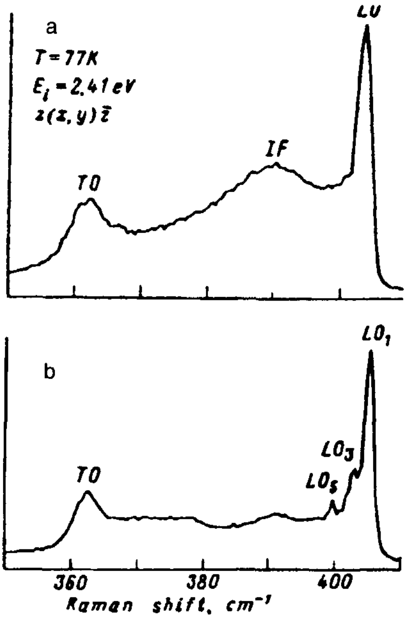

FIG. 1. Raman-scattering spectra in the AlAs frequency region. a—$(GaAs)_7(AlAs)_7$ superlattices with rough interfaces; b—$(GaAs)_9(AlAs)_9$ superlattices with sharper interfaces.

trum because of a violation of wave-vector conservation. This circumstance is a serious complication in efforts to observe and correctly interpret the peaks of confined optical phonons. Furthermore, a disorder of the interfaces leads to significant changes in the frequencies of the confined LO phonons.[8] In this case, the experimental values of the frequencies of confined optical phonons evidently cannot be used to construct the dispersion of the bulk material.

For the experiments we selected GaAs/AlAs superlattice samples of the highest quality. A criterion in choosing samples was that there be no interface feature in the Raman-scattering spectra (in either the GaAs region or the AlAs region). It was thus possible to spectrally resolve the peaks of confined LO phonons of AlAs (Fig. 1b).

We studied the following samples, which met this criterion in terms of interface quality: $(GaAs)_{12}(AlAs)_{12}$, $(GaAs)_{10}(AlAs)_{10}$, $(GaAs)_9(AlAs)_9$, $(GaAs)_{12}(AlAs)_6$, and $(GaAs)_9(AlAs)_5$. All the test samples were grown at the Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, by molecular beam epitaxy on GaAs substrates in the (001) orientation. The thickness of each layer was monitored during the growth by detecting oscillations in the intensity of the specular reflection in reflection high-energy electron diffraction. Raman-scattering spectra were recorded by a Jobin Yvon U1000 spectrometer, with excitation by the light from an argon laser with a wavelength of 514.5 nm at a temperature of 77 K in the configurations $z(x,y)\bar{z}$ and $z(x,x)\bar{z}$ [the z axis runs parallel to the (001) growth direction of the superlattice, while the x and y axes are along the (100) and (010) directions, respectively]. In all these samples, LO_1, LO_3, and LO_5 phonons confined in the AlAs layers were spectrally resolved in the $z(x,y)\bar{z}$ configuration (the spectra of all the superlattices are similar to that shown in Fig. 1b). Furthermore, in the parallel geometry $z(x,x)\bar{z}$ we observed peaks

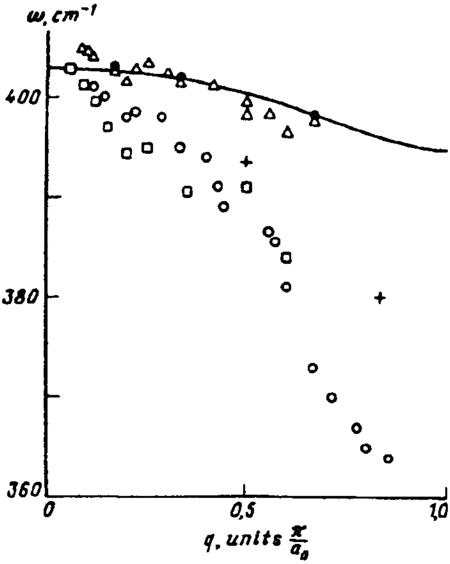

FIG. 2. Dispersion of LO phonons of bulk AlAs. Δ—Data of the present study; ●—data of Ref. 5; ○, □—data of Refs. 6 and 7, respectively; +—calculated frequencies of confined AlAs LO phonons in a $(GaAs)_5(AlAs)_5$ superlattice. The solid curve is the result of a microscopic calculation.[3]

of four confined LO_2 phonons; for a $(GaAs)_{12}(AlAs)_6$ sample, we also observed an LO_4 peak.

Working from these results, we can construct the dispersion of the LO phonons of bulk AlAs. For this purpose, we compare the experimental frequency of a confined phonon, LO_m, with the wave vector found from relation (1). It follows from an analysis of the complex dispersion relation of GaAs (Ref. 9) that at the frequencies of AlAs LO phonons ($\sim 400\ \mathrm{cm}^{-1}$) the relation $(\mathrm{Im}(k))^{-1} < 1$ Å holds [$\mathrm{Im}(k)$ is the imaginary part of the wave vector, which describes the attenuation of AlAs phonons in the GaAs layers]. We accordingly set $\delta = 0$ in relation (1). [In an analysis of the penetration of a GaAs LO phonon into the AlAs layer we have $(\mathrm{Im}(k))^{-1} \approx 3$ Å ; correspondingly, the parameter δ is usually assumed to be one monolayer.]

The triangles in Fig. 2 show the dispersion of the LO phonons of bulk AlAs constructed from our data. Also shown in this figure are experimental data from Ref. 5 and also from Refs. 6 and 7. The solid curve is the result of a microscopic *ab initio* calculation of the dispersion of LO phonons of AlAs (Ref. 3). Our data, which are quite different from the data of Refs. 6 and 7, agree well with the data of Ref. 5 and also with the theoretical dispersion. The experimental points do deviate slightly from the theoretical dispersion, perhaps because of imperfections of the superlattices. As has been mentioned previously,[8] such imperfections reduce the frequency of confined LO phonons in AlAs. To illustrate this point, we show in Fig. 2 (by the plus signs) calculated frequencies of confined AlAs LO phonons in a $(GaAs)_5(AlAs)_5$ superlattice. In this superlattice the material over two monolayers at each interface corresponds to the solid solution $Ga_{0.5}Al_{0.5}As$ (Ref. 8). It can be seen from Fig. 2 that these values approach the data of Refs. 6 and 7.

We also studied the spectrum of AlAs phonons in several $(GaAs)_8(AlAs)_n$ superlat-

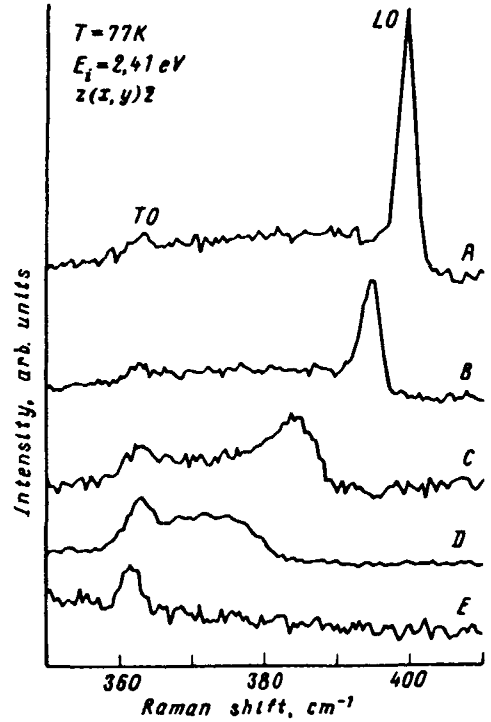

FIG. 3. Raman-scattering spectra of $(GaAs)_8$ $(AlAs)_n$ superlattice samples with $n=3,2,1$, and 0.5 monolayers (A,B,C, and D, respectively). Spectrum E is that of a $Ga_{1-x}Al_xAs$ solid solution with $x\approx 0.01$.

tices with $n=3,2,1$, and 0.5 monolayers; and the solid solution $Ga_{1-x}Al_xAs$, with $x\sim 0.01$. In these spectra we can trace the evolution of an LO phonon confined in an AlAs layer into a local phonon of an Al atom in GaAs. Figure 3 shows Raman-scattering spectra of these samples. It can be seen from this figure that in superlattices in which the AlAs layers have thicknesses of 3 and 2 monolayers the frequency of an LO phonon lies within the frequency interval of the LO phonons of AlAs. In other words, a phonon in these superlattices can still be thought of as a confined phonon in an AlAs layer. (However, data on its frequency in these superlattices cannot be used to construct the dispersion of bulk AlAs, since the disorder of the interfaces in superlattices with ultrathin layers is very substantial.) With decreasing thickness of the AlAs layers, the frequency of LO phonon decreases, approaching the frequency of a local phonon of an Al atom in GaAs, which we observed in the spectrum of a solid solution with a low Al concentration (about 1%).

In summary, we have reported an experimental Raman-scattering study of confined LO phonons in the AlAs layers of GaAs/AlAs superlattices. The results confirm a microscopic *ab initio* calculation on the dispersion of AlAs LO phonons. We have also studied the spectrum of AlAs phonons in superlattices with ultrathin AlAs layers. This study has made it possible to observe the evolution of a confined phonon in AlAs layers into a local vibration of an Al atom in GaAs.

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