Raman scattering by *LO* phonons in GaAs/AlAs superlattices with ultrathin AlAs layers

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A study of Raman scattering by longitudinal optical phonons in GaAs/AlAs superlattices with ultrathin AlAs layers has made it possible to observe scattering by odd and even localized *LO* phonons simultaneously.
An explanation for this result is proposed: a mechanism of Raman scattering due to defects.

Since the frequency ranges of optical phonons in GaAs and AlAs do not overlap, optical phonons in GaAs/AlAs superlattices are localized in the layers of one of the materials.[1] The depth to which a GaAs *LO* phonon penetrates into an AlAs layer is ~1 monolayer.[2,3] This localization of optical phonons is manifested in Raman scattering spectra as a series of peaks, whose frequencies correspond to the frequencies of the *LO* phonons of bulk GaAs at wave-vector values

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

where a_0 is the thickness of a GaAs monolayer, and n is the number of monolayers.[1] In GaAs/AlAs superlattices grown in the (001) direction, a scattering by odd localized phonons ($m=1,3,5,...$) is observed in the $z(x,y)\bar{z}$ geometry under nonresonant conditions. A scattering by even phonons is possible in the $z(x,x)\bar{z}$ parallel geometry under resonant conditions.[1,2] In the present letter we are reporting the observation, under nonresonant conditions, of a scattering by odd and even localized *LO* phonons simultaneously in GaAs/AlAs superlattices with ultrathin AlAs layers.

We have used the Raman scattering method to study samples of $(\mathrm{GaAs})_n(\mathrm{AlAs})_l$ superlattices (n and l are the numbers of monolayers) with $n=8$ in all cases and with $l=3,2,1$, and 0.5 for samples *A*, *B*, *C*, and *D*, respectively. All the samples were grown by molecular beam epitaxy on apparatus developed in the Institute of Semiconductor Physics on GaAs(001) substrates. The layer thicknesses were monitored during the growth by detecting oscillations in the intensity of the specular reflection in reflection high-energy electron diffraction. The Raman spectra were recorded on a *U*-1000 spectrometer during excitation by light from an argon laser with a wavelength of 514.5 nm at 77 K in a $z(x,y)\bar{z}$ quasi-back-scattering geometry.

Figure 1 shows Raman spectra of samples *A*, *B*, *C*, and *D*. The spectra of samples *A* and *B* are characteristic of scattering by localized optical phonons in the $z(x,y)\bar{z}$ geometry under nonresonant conditions.[1] These spectra contain peaks corresponding

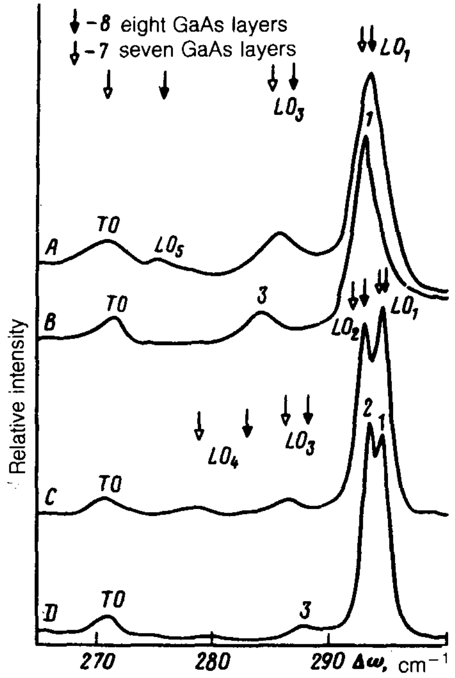

FIG. 1. Raman spectra of samples *A–D*. The arrows show the calculated frequencies of localized *LO* phonons.

to scattering by LO_1, LO_3, and LO_5 odd localized phonons. The spectra of samples *C* and *D* are quite different from those of samples *A* and *B*. They have peaks corresponding to both odd and even localized *LO* phonons.

Figure 2 shows dispersion curves for *LO* phonons in $(GaAs)_8(AlAs)_1$ and $(GaAs)_8(AlAs)_3$ superlattices according to calculations from a linear-chain model. For the $(GaAs)_8(AlAs)_1$ superlattice, the calculations were carried out with the help of the expression derived in Ref. 4 for the dispersion specifically for superlattices containing ultrathin layers (1 or 2 monolayers) of the material of one type, with no restriction on the thickness of the layers of the other material. The calculations used the dispersion derived for *LO* phonons of bulk GaAs and AlAs in Ref. 5, which agree well with experimental data.

We see in Fig. 1 that the experimental frequencies of the *LO* phonons in samples *A–D* agree well with the theoretical frequencies derived from the dispersion curves in Fig. 2. For samples *B* and *C*, however, the best agreement is reached when the thickness of the GaAs layer is seven monolayers. For sample *B* this value of the thickness is supported by data on Raman scattering by folded acoustic phonons. These results make it possible to determine the overall period of the superlattice:[1-3] nine monolayers.

Two suggestions have been offered in an effort to explain the appearance of peaks corresponding to even localized *LO* phonons in the Raman spectra. First, it was suggested that if the thickness of the AlAs layers is only 1 monolayer, then the GaAs *LO* phonons can penetrate through such thin barriers and become delocalized. Indeed,

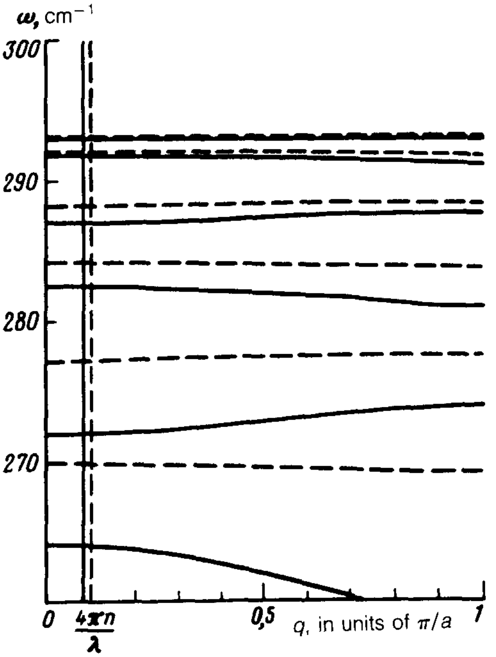

FIG. 2. Dispersion curves of *LO* phonons in $(GaAs)_8(AlAs)_1$ superlattice (solid lines) and $(GaAs)_8(AlAs)_3$ superlattice (dashed lines).

we see in Fig. 2 that the branches of the dispersion curve for *LO* phonons in the $(GaAs)_8(AlAs)_1$ superlattice are tilted somewhat, implying that there is something less than a strict localization of the phonons in the GaAs layers. On the other hand, this tilt is only slight, and the atomic displacements which we calculated in the linear-chain model for the value of the wave vector $q=4\pi n/\lambda$, corresponding to the backscattering geometry (λ is the wavelength of the incident light, and n is the refractive index), differ only slightly from those corresponding to the case of strict localization. The intensity ratio of the LO_1 and LO_2 peaks found on this basis is $\sim 10^2$ (we used the calculation method of Ref. 6), at odds with the experimental data. (Figure 1 shows that experimentally these peaks are of approximately the same intensity.)

The second suggestion is based on the idea that even in the highest-quality superlattices there will always be fluctuations in the layer thickness with a magnitude ~ 1 monolayer, and if the thickness of an AlAs layer is only 1 monolayer, then this layer will be missing completely in certain regions. A disorder arises in the system. This disorder does not change the overall vibrational spectrum of the superlattice, but it does have the consequence that scattering processes involving a violation of wave-vector conservation become possible along with processes which do conserve the wave vector and which lead to a scattering by odd localized *LO* phonons. In this case the Raman spectrum should have a component proportional to the density of phonon states in the superlattice. We thus link the appearance of peaks corresponding to even localized *LO* phonons with scattering processes in which wave-vector conservation is violated. In superlattices in which the thickness of the AlAs layer amounts to two or more monolayers, fluctuations in the thickness on the order of one monolayer are far less likely to lead to the formation of regions in which the AlAs layer is missing. We thus see why the Raman spectra of superlattices containing two and three AlAs monolayers do not exhibit the peaks corresponding to even localized *LO* phonons.

In summary, we have reported the simultaneous observation of a nonresonant Raman scattering by odd and even localized *LO* phonons in GaAs/AlAs superlattices with ultrathin AlAs layers grown in the (001) direction. A possible explanation for this fact is that in such superlattices defects (i.e., regions lacking the AlAs layer) intensify Raman scattering processes in which the wave vector is not conserved, so peaks corresponding to even localized *LO* phonons appear in the spectra.

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