Resonant penetration of optical vibrational modes in ultrathin $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices

Yu. A. Pusep

Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

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The interaction of optical vibrational modes localized in layers of $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices has been studied by IR Fourier spectroscopy and Raman-scattering spectroscopy. A resonant penetration of transverse, optical, quantized vibrational modes into a barrier has been observed for the first time.

The optical frequencies in GaAs and AlAs lie in different ranges because of the large difference between the masses of the cations. Consequently, optical vibrations in GaAs/AlAs superlattices are localized in the corresponding layers. The AlAs layers act as barriers to the GaAs vibrations, and the GaAs layers do the same to the AlAs vibrations.

The vibrational spectra of $Al_xGa_{1-x}As$ solid solutions exhibit a two-mode behavior:[1] One mode, with frequencies close to those of AlAs, is called an "AlAs-like" mode, while another mode (the "GaAs-like" mode) lies near the optical frequencies of GaAs. As a result, the dispersion curves of the optical vibrations in neighboring layers of a $GaAs/Al_xGa_{1-x}As$ superlattice may overlap. The vibrational modes localized in the GaAs and $Al_xGa_{1-x}As$ layers which have the same frequencies are therefore called "delocalized." "Penetrating" optical modes of this sort obviously cannot interact with light because their effective wave number is large. They accordingly cannot be seen in optical spectra (IR or Raman spectra).

We have used IR Fourier spectroscopy to study the different behavior of the optical vibrational modes in $GaAs/Al_xGa_{1-x}As$ superlattices. In this case, it is simpler to satisfy the conditions for resonances between quantized modes in the different layers because the dispersion of the transverse optical (TO) phonons, which are active in the IR spectra, is slight in comparison with the dispersion of longitudinal optical (LO) phonons, which are active in Raman scattering.

To study the resonant penetration of optical vibrational modes, we measured the reflection spectra of $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices, where n is the number of monolayers in the layers of the superlattice. These measurements were carried out in the frequency range of TO phonons of GaAs at room temperature. At room temperature, it is a relatively simple matter to satisfy the conditions for resonances between modes localized in different layers.

The reflection spectra were recorded with the help of a Bomen DA-8 Fourier spectrometer with a spectral resolution of 1 cm^{-1}. To determine the frequencies of the quantized modes accurately, we analyzed spectra of the first derivative of the reflec-

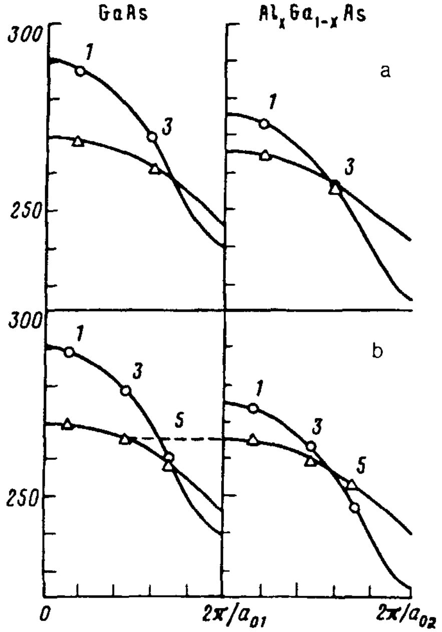

FIG. 1. Spectrum of optical vibrational modes of $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices which are active in the IR. a—$n=4$; b—$n=6$. Circles—LO modes; triangles—TO modes.

tion, using the method described in Ref. 2. The frequencies of the optical quantized modes were found by fitting theoretical spectra of the derivative of the reflection to the experimental spectra. The theoretical reflection spectra were calculated by the method described in Ref. 3.

The $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices studied in these experiments, with ultrathin layers, were grown by molecular beam epitaxy on GaAs (100) substrates. In order to see the quantized modes in a single layer at a good energy resolution, we used superlattices with $n=4$ and $n=6$. The total thickness of the superlattices was 8960 Å (400 repetitions) for the samples with $n=4$ and 6720 Å (200 repetitions) for those with $n=6$. Under these conditions it was possible to observe a substantial reflection due to the optical vibrational modes localized in the layers.

Figure 1 shows a spectrum of optical modes of these superlattices in the region of GaAs vibrations. The frequencies of GaAs and $Al_{0.5}Ga_{0.5}As$ near the Γ point of the Brillouin zone were taken from Ref. 1; the dispersion for TO and LO phonons of GaAs and AlAs was taken from Refs. 2 and 4, respectively. Dispersion curves for the GaAs-like vibrations in $Al_{0.5}Ga_{0.5}As$ were found by shifting the dispersion for the GaAs optical vibrations by amounts corresponding to centrally symmetric optical phonons in $Al_{0.5}Ga_{0.5}As$ (Ref. 1). To determine the effective wave numbers of the quantized modes (q_m) we used the relation[5]

$$q_m=2\pi\, m/(n+1)a_{01,2} ,$$

where m is the index of the quantized mode, and $a_{01,2}$ are the lattice constants of GaAs and $Al_{0.5}Ga_{0.5}As$, respectively.

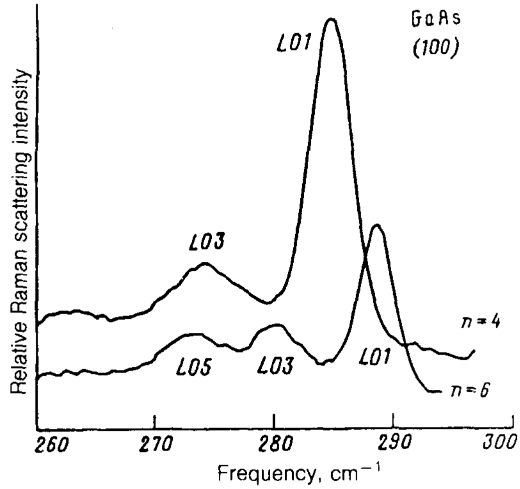

FIG. 2. Intensity of Raman scattering (I_R) in the region of LO vibrations of GaAs in $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices.

We see in Fig. 1 that in the superlattices with $n=4$ all eight optical quantized modes (four LO modes and four TO modes) are resolved well in terms of energy and should be observable in the spectra (IR or Raman). In the $n=6$ case, on the other hand, all modes are localized except two: the TO_3 mode in the GaAs layer overlaps with the GaAs-like TO_1 mode in the $Al_{0.5}Ga_{0.5}As$ layer (as shown by the dashed line in Fig. 1), forming a single "penetrating" mode. These two quantized modes thus cannot be seen in the experimental spectra.

To verify that the LO modes were localized, as expected on the basis of Fig. 1, we measured spectra of the Raman scattering by LO phonons of GaAs. Figure 2 shows Raman scattering spectra corresponding to the scattering of all LO modes localized in the GaAs layers. We did not observe Raman spectra in the region of optical vibrations of $Al_{0.5}Ga_{0.5}As$ because of their low intensity.

Figure 3 shows spectra of the derivative of the reflection for two samples with $n=4$ and $n=6$; these spectra were recorded in the region of GaAs TO vibrations. Two spectral lines are observed in the sample with $n=4$: an intense doublet near 265 cm^{-1}, which is due to a GaAs-like TO_1 mode and a GaAs TO_1 mode, which are localized in $Al_{0.5}Ga_{0.5}As$ and GaAs layers, respectively. The TO vibrations of the substrate also contribute at the frequency of 267 cm^{-1}, but the intensity of the reflection from the substrate is comparatively low, because of the large thickness of the superlattice. The second line, observed near 255 cm^{-1}, is due to a GaAs-like TO_3 quantized mode (localized in the $Al_{0.5}Ga_{0.5}As$ layers) and possibly a GaAs TO_3 mode (localized in the GaAs layers). This line was not intense enough to support a reliable determination of the frequencies of the GaAs and GaAs-like vibrational TO_3 modes.

In the spectrum of the derivative of the reflection of a sample with $n=6$ we found only a single intense line, at 266 cm^{-1}. This line is due to the fundamental GaAs TO_1 mode. In agreement with Fig. 1, the quantized GaAs-like TO_1 and GaAs TO_3 modes overlap in this case, *forming a single penetrating mode, which is inactive in the* IR spectra. The slight knee which can be seen on the low-frequency side of the TO_1

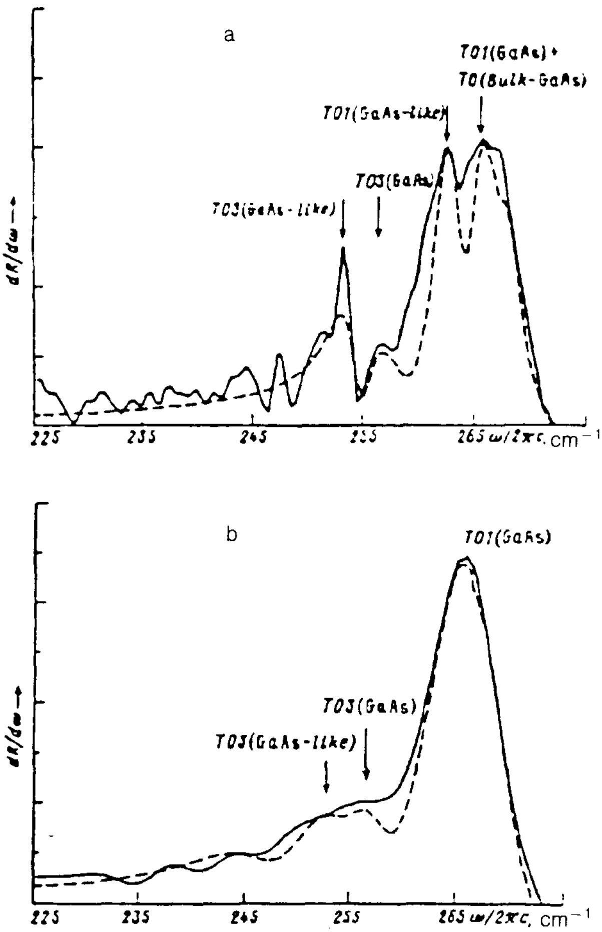

FIG. 3. Spectrum of the derivative of the reflection in the region of TO vibrations of GaAs in $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices. a—$n=4$; b—$n=6$.

line (Fig. 3) indicates a possible contribution of GaAs TO_3 and GaAs-like TO_1 modes, which overlap to some extent. Consequently, their "localized part" can interact with the light.

In summary, this study of optical vibrational modes in $(GaAs)_n(Al_{0.5}Ga_{0.5}As)_n$ superlattices has shown that the overlap of the dispersion of transverse optical phonons in GaAs with that in $Al_{0.5}Ga_{0.5}As$ leads to the appearance of some new penetrating modes, which do not interact with the light because of their large effective wave number. This effect is the reason why quantized modes with identical frequencies localized in neighboring layers disappear from the optical spectra.

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