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LETTER TO THE EDITOR

Lattice vibrations of ultrathin-layer $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ superlattices

Yu A Pusep† and A I Toropov‡

† I. Physikalisches Institut der RWTH, D-5100 Aachen 1, Federal Republic of Germany
‡ Institute of Semiconductor Physics, 630090 Novosibirsk, Russia

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Abstract. We present experimental results obtained by far-infrared reflection spectroscopy in the investigation of the optical phonons in ultrathin-layer $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ superlattices (UTSLs). It was found that the frequencies of confined optical phonons in UTSLs are much closer to the frequency of the corresponding bulk phonon at the centre of the Brillouin zone than in SLs with sufficiently thick layers. The influence of the barrier thickness was also studied.

In recent years UTSLs have been extensively studied in order to obtain information about the influence of interatomic interactions on the vibrational properties of SLs. Raman scattering has been used to analyse the vibrational properties of such SLs as allowed one to investigate the longitudinal optical (LO) phonons [1,2]. Data on tranverse optical (TO) phonons in $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ UTSLs have been recently obtained by infrared measurements [3]. It has been found that in the case of a layer thickness of about one monolayer it is not possible to explain the deviation of the observed values of confined LO phonon frequencies from calculated ones by means of the linear-chain model [2]. It was pointed out that microscopic calculations should be used in this case. On the other hand, good agreement with a linear-chain model was found in [1]. The experimental data for both cases are in good correspondence; we suppose that the difference between the experimental data and the linear-chain model results is caused by using different parameters in the model used for the calculations.

To avoid making an inaccurate comparison with the theoretical models, we compared the frequencies of optical phonons of UTSLs with the frequencies of SLs with thick layers. We have used far-infrared reflectivity measurements to study the LO and TO vibrations in $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ UTSLs. As has been pointed out in [4], with the help of infrared spectroscopy it is possible to observe the odd confined TO phonons in SLs. The LO superlattice vibrational modes observed in infrared spectra correspond to the superposition of all confined LO phonons, which is caused by the Coulomb coupling between them. Thus the change of the frequencies of LO confined phonons in UTSLs should be accumulated. This effect provides us with the possibility of observing very small shifts of the frequencies of LO phonons in the UTSLs.

We compared the values of frequencies of the first confined TO modes and LO superlattice modes of UTSLs with the confined TO and LO superlattice modes with corresponding wavenumbers of SLs with thick enough layers (10 and 17 monolayers).

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It has been found that the frequencies of TO modes in UTSLs are changed, while no significant shift of the LO modes was observed. The SLs under investigation were grown by molecular beam epitaxy on (100) GaAs substrates. We investigated the SLs with $n = m$, where n and m are the number of monolayers of GaAs and AlAs layers respectively. The SL with $n \gg m$ has been used to study the influence of the barrier thickness on the phonons in AlAs layers. The layer structure was repeated 50 times for all SLs under investigation except the SL with $m = 2$. In the latter case the number of periods was 100. In order to obtain more pronounced reflection spectra of the SLs, highly doped substrates (with a doping level of about 2×10^{18} cm^{-3}) have been used.

To study the SL vibrational modes we measured the reflection at oblique incidence ($\theta = 70°$) in p-polarized light. In this case two components of electric polarization of the light should be taken into account: the first one is directed in the plane of the layers and the second one is normal to the layers. In the layered structure these different components of electric polarization of the light are coupled with the difference lattice vibrations [5]. The parallel component is coupled with TO vibrations while the normal one has a resonance at the frequency of LO vibrations of SL. Thus it is possible to investigate both TO and LO vibrations of SLs by means of infrared spectroscopy.

We present the results of the investigations of the optical vibrations of AlAs layers in $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ UTSLs. In principle, the behaviour of GaAs vibrational modes of these SLs was the same, although it was rather difficult to measure the values of the frequencies of these modes accurately because of the vicinity of the bulk optical vibrations of the GaAs substrate. The theoretical treatment presented in [6] has been used to analyse the experimental reflection spectra of the SLs.

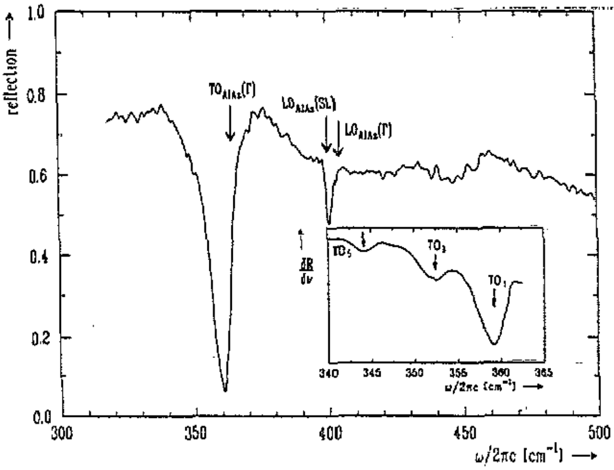

Figure 1. p-polarized reflectivity at $T = 80$ K of a $(\mathrm{GaAs})_5(\mathrm{AlAs})_5$ SL in the spectral range of AlAs vibrations. The inset shows the derivative of the reflection spectra of a $(\mathrm{GaAs})_{10}(\mathrm{AlAs})_{10}$ SL in the spectral range of TO vibrations of AlAs. Arrows show the positions of bulk optical phonons of AlAs (at the centre of the Brillouin zone) and the positions of LO superlattice and TO confined modes.

A typical reflection spectrum of $(\mathrm{GaAs})_n(\mathrm{AlAs})_m$ SLs in the spectral range of

AlAs lattice vibrations is plotted in figure 1. The inset shows the derivative of the reflection of a $(GaAs)_{10}(AlAs)_{10}$ SL, which allowed us to measure the frequencies of confined TO modes very accurately. The first confined TO_1 modes of UTSLs with different layer thicknesses are presented in figure 2. The confined TO modes of higher index for SLs with $n = m = 10$ and 17, which are in good agreement with the dispersion of bulk TO phonons of AlAs, are shown in figure 2 as well. The effective wavenumbers of the confined phonon modes were calculated by means of an expression that takes into account the real position of the mode nodes:

$$K_j = 2\pi j/(m+1)a_0$$

where m is the layer thickness in monolayers, j is the mode index and a_0 is the lattice constant.

As shown in figure 2, the frequencies of fundamental TO vibrations measured on UTSLs differ from the frequencies of high-index confined TO modes with corresponding wavenumbers obtained for SLs with thick layers. This difference reaches a value of about 15 cm^{-1} in the UTSL with a thickness of AlAs layers of two monolayers.

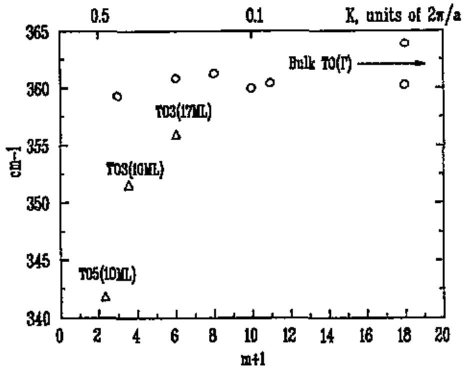

Figure 2. Frequencies of the first AlAs TO confined modes (open circles) and high-index AlAs TO confined modes (triangles) plotted as functions of $m+1$ and corresponding wavenumbers K.

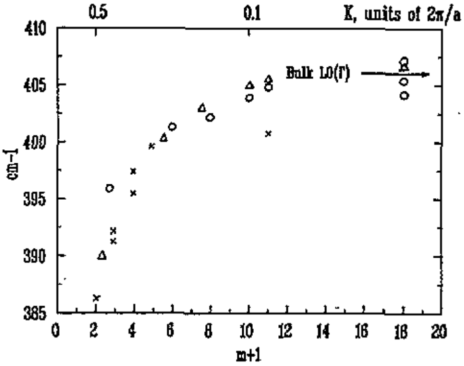

Figure 3. Frequencies of the AlAs LO superlattice modes (open circles) plotted as functions of $m+1$ and corresponding wavenumbers K. Triangles show the calculated LO superlattice modes of superlattices with thick layers ($m = n = 10$ and 17). Crosses show the Raman data from [8].

The experimental results for LO superlattice modes are presented in figure 3. The calculated values of frequencies of LO superlattice modes were obtained by means of the dispersion of LO phonons of AlAs measured in [1,7] and TO phonons after [4] which were obtained for SLs with thick AlAs layers. These data were put into the dielectric response function tensor of the SL [6], and thus the frequencies of LO superlattice modes were calculated. It should be mentioned that in the case of UTSL with a thickness of AlAs layers of two monolayers, the LO superlattice mode is the real confined LO phonon mode. The data presented in figure 3 show very little difference between the frequencies of confined phonons in the USTLs and in the thick-layer SLs, which can be explained by experimental error. The Raman data on AlAs LO phonons obtained for the UTSLs $(GaAs)_n(AlAs)_m$ in [8] are presented in figure 3 too.

The different behaviour of TO and LO vibrational modes observed in UTSLs can be explained in such a way. The infrared spectra for oblique incidence of the light present TO vibrational modes with electric polarization directed in the plane of the layers and LO modes polarized normal to the layers. The electric field associated with the TO modes is conserved at the interfaces and therefore penetrates into the barrier farther than the electric field associated with the LO ones. Thus the LO vibrations observed in infrared spectra are more strongly localized and are not so sensitive to the barrier thickness as the TO ones.

As has been pointed out in [1], the origin of the shift of confined phonons in UTSLs consists in the coupling of the optical vibrations of the identical layers through the barriers; because of this the frequencies of confined phonons of UTSLs are closer to the bulk phonons. However, we observed the same shift of optical vibrations of ultrathin AlAs ($m = 2$) layers for SL with very thick GaAs layers ($n = 25$). This means that size effects of the individual AlAs layer are also responsible for the shift of the optical vibrations observed in UTSLs.

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