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Abstract. We present a study of the anisotropy of zone-centre optical phonons in (GaAs)$_n$/(AlAs)$_m$ superlattices by Fourier-transform infrared spectroscopy. We show that the reflection spectra of semiconductor superlattices taken at off-normal incidence of the light contain information about both confined phonons propagating normal to the layers and interface phonons propagating in the plane of the layers. The frequencies of confined and interface modes obtained from reflection spectra are compared with those calculated by dielectric continuum theory.

Layered systems such as superlattices (SLs) are anisotropic crystals with the axis directed parallel to the growth direction. For this reason the vibrational spectrum of SLs reveals the angular dispersion; according to theoretical calculations (see for instance [1–3]) the frequencies of optical phonons propagating in the plane of the layers differ from the frequencies of phonons with the wavevector normal to the layers. This angular dispersion strongly depends on the total dipole moment of relevant modes; thus only odd modes confined in the layers of the SL which have a non-zero dipole moment present angular dispersion while even modes are dispersionless.

By means of infrared spectroscopy it is possible to measure directly the oscillator strengths of confined modes (this is not possible by Raman spectroscopy) which are proportional to their dipole moments and hence to study the angular dispersion of confined modes in SLs.

The first application of FTIR spectroscopy to the problem of investigation of anisotropy in GaAs/AlAs SLs was described in [4]; thick-layer SLs were studied, and neither confined nor interface modes were found.

In this paper we show that the analysis of reflection spectra of SLs taken with off-normal incidence of the light makes it possible to measure the frequencies of modes propagating normal to the layers (confined modes) and in the plane of the layers (interface modes) and hence to obtain the angular dispersion of the relevant optical vibrations. The interface vibrational modes are more sensitive to the quality of interfaces than are the confined modes. Thus FTIR spectroscopy presents the possibility of studying the interface modes more easily than by Raman scattering (in the usually used back-scattering geometry the interface modes are forbidden) [5] and hence of characterizing the interface quality more accurately.

By using the simple dielectric continuum model [6–8] with the SL described as a uniaxial medium it is possible to calculate optical modes with arbitrary wavevectors. In the long-wavelength approximation (when $\lambda \gg d$, where d is the SL period and λ is the wavelength

of the light) the dielectric response function can be presented as a tensor:

$$\varepsilon_{sl}(\omega) = \begin{bmatrix} \varepsilon_x(\omega) & 0 & 0 \\ 0 & \varepsilon_y(\omega) & 0 \\ 0 & 0 & \varepsilon_z(\omega) \end{bmatrix} \tag{1}$$

where the z axis is taken normal to the plane of layers.

In the short-period SLs when the effect of quantization of optical phonons plays an important role, the microscopic theory gives the components of the dielectric function tensor [9]:

$$\varepsilon_x(\omega) = \varepsilon_y(\omega) = \varepsilon_{x\infty}\left(1 - \sum_{\mu,\lambda} \frac{R_{t\mu}^{(\lambda)}}{\omega^2 - \omega_{t\mu}^{(\lambda)2} + i\Gamma_{x\mu}^{(\lambda)}\omega}\right) \tag{2}$$

$$\frac{1}{\varepsilon_z}(\omega) = \frac{1}{\varepsilon_{z\infty}}\left(1 + \sum_{\mu,\lambda} \frac{R_{l\mu}^{(\lambda)}}{\omega^2 - \omega_{l\mu}^{(\lambda)2} + i\Gamma_{z\mu}^{(\lambda)}\omega}\right) \tag{3}$$

where $\omega_{t\mu}^{(\lambda)}$ and $\omega_{l\mu}^{(\lambda)}$ are the frequencies of the confined TO and LO phonons in layer λ ($\lambda = 1$ for GaAs and $\lambda = 2$ for AlAs), μ is the index of the mode, $R_{t\mu}^{(\lambda)}$ and $R_{l\mu}^{(\lambda)}$ are their oscillator strengths, $\varepsilon_{x\infty}$ and $\varepsilon_{z\infty}$ are the principal components of the high-frequency dielectric function, and $\Gamma_{x\mu}^{(\lambda)}$ and $\Gamma_{z\mu}^{(\lambda)}$ are the damping constants for the corresponding modes.

The poles of $\varepsilon_{x,y}(\omega)$ and the zeros of $\varepsilon_z(\omega)$ give the frequencies of confined TO$_{x,y}$ and LO$_z$ modes, respectively, while the zeroes of $\varepsilon_{x,y}(\omega)$ and the poles of $\varepsilon_z(\omega)$ give the frequencies of the interface LO$_{x,y}$ and TO$_z$ vibrational modes, respectively. The substitution of experimental values of confined mode frequencies $\omega_{t(l)\mu}^{(\lambda)}$ and their oscillator strengths $R_{t(l)\mu}^{(\lambda)}$ into (2) and (3) makes it possible to calculate the frequencies of interface modes.

Application of the Maxwell equations to the SL gives the expressions for propagation of s- and p-polarized light:

$$(q_x^2 + q_z^2)/\varepsilon_x = \omega^2/c^2 \tag{4}$$

$$q_z^2/\varepsilon_x + q_x^2/\varepsilon_z^2 = \omega^2/c^2 \tag{5}$$

where q_x and q_z are the x and z components, respectively, of the wavevector and c is the velocity of light. In the non-retarded limit, equations (4) and (5) give

$$\varepsilon_x^{-1} = 0 \tag{6}$$

$$\varepsilon_z/\varepsilon_x = -\tan^2\vartheta \tag{7}$$

where $\tan\vartheta = q_x/q_z$ and ϑ is the angle of propagation of vibrational modes measured from the z axis. The first equation (6) describes the propagation of s-polarized modes while the second equation (7) is valid in the case of p-polarization. It follows from (6) and (7) that the p-polarized modes are isotopic whilst the p-polarized modes reveal angular dispersion.

The measurement of infrared spectra taken with off-normal incidence of p-polarized light makes it possible to probe both the ε_x and the ε_z components of the dielectric response function tensor of the SL. Then the angular dispersion of the p-polarized optical modes may be calculated according to (7) with ε_x and ε_z as presented by equations (2), (3). The values of frequencies of the confined modes TO$_x$ and LO$_z$ and their oscillator strengths $R_{t\mu}^{(\lambda)}$ and

$R_{1\mu}^{(\lambda)}$ which were indispensable for calculating ε_x and ε_z have been obtained from the fitting of the theoretical reflection spectra to the experimental spectra. The reflection spectra of SLs were calculated using the dielectric function tensor (1) as in [10].

According to the symmetry considerations the $(GaAs)_n/(AlAs)_m$ SLs (where n and m are the numbers of monolayers) with the axis parallel to the [100] direction are characterized by the basic space group D_{2d}. Thus it turned out that the transverse optical vibrational modes $TO_{x,y}$ and TO_z considered above are infrared active modes of E and B_2 symmetries, respectively. In addition, LO_z modes are infrared active owing to the Berreman [11] effect.

The samples under investigation have been grown by MBE on doped GaAs (100) substrates. The use of doped substrates enhanced the intensity of spectral lines caused by the vibrational modes of SL and eliminated the interference due to the total thickness of the sample. The reflection spectra have been recorded with a Bruker IFS-113V Fourier-transform infrared spectrometer at the temperature $T = 80$ K.

In order to avoid the influence of the GaAs substrate we analysed only the AlAs-like vibrations. The reflection spectra of some SLs measured in the spectral range of AlAs optical phonons with p-polarized light are plotted in figure 1. The results of the fitting of the calculated spectra to the experimental spectra are plotted as dashed curves. The values of the frequencies of confined optical modes and their oscillator strengths obtained by the fitting procedure have been used to calculate the angular dispersion of confined modes in these SLs. The oscillator strengths deduced for the confined optical modes are collected in table 1.
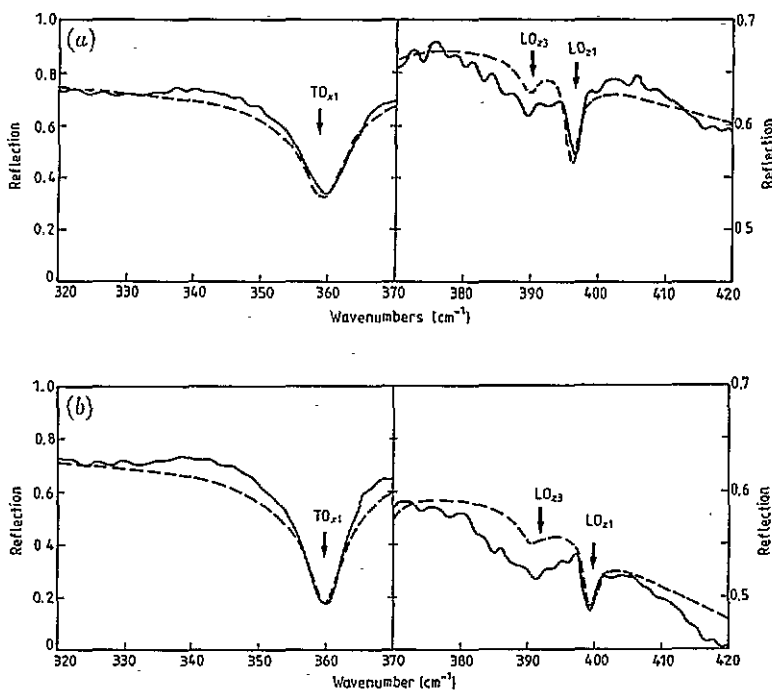

Figure 1. The spectra of p-polarized reflection measured in the range of AlAs optical vibrations at $T = 80$ K for (a) $(GaAs)_8/(AlAs)_3$ and (b) $(GaAs)_{10}/(AlAs)_4$: - - - -, calculated reflection spectra. The subscripts in the mode labels denote their electric polarizations and quantum numbers, respectively.

Table 1. The oscillator strengths of optical confined modes measured in $(GaAs)_n/(AlAs)_m$ sls by FTIR at the temperature $T = 80$ K.

Samples	$R_{11}^{(2)}$ (cm^{-2})	$R_{13}^{(2)}$ (cm^{-2})	$R_{tl}^{(2)}$ (cm^{-2})
$(GaAs)_8/(AlAs)_3$	25 000	10 000	12 000
$(GaAs)_{10}/(AlAs)_4$	20 000	10 000	10 000
$(GaAs)_5/(AlAs)_5$ $N = 1 \times 10^{17}$ cm^{-3}	50 000	—	50 000
$(GaAs)_5/(AlAs)_5$ $N = 2 \times 10^{18}$ cm^{-3}	10 000	—	27 000

The calculated angular dispersion of optical phonons in different samples with the parameters measured by means of infrared reflection spectra are displayed in figure 2. It should be pointed out that the values of oscillator strengths of confined modes obtained from the fitting of calculated reflection spectra to the experimental spectra strongly depend on the structural quality of layers and interfaces and as a result the frequencies of the interface modes TO_z and LO_x calculated with the experimental values of oscillator strengths depend on these factors as well.
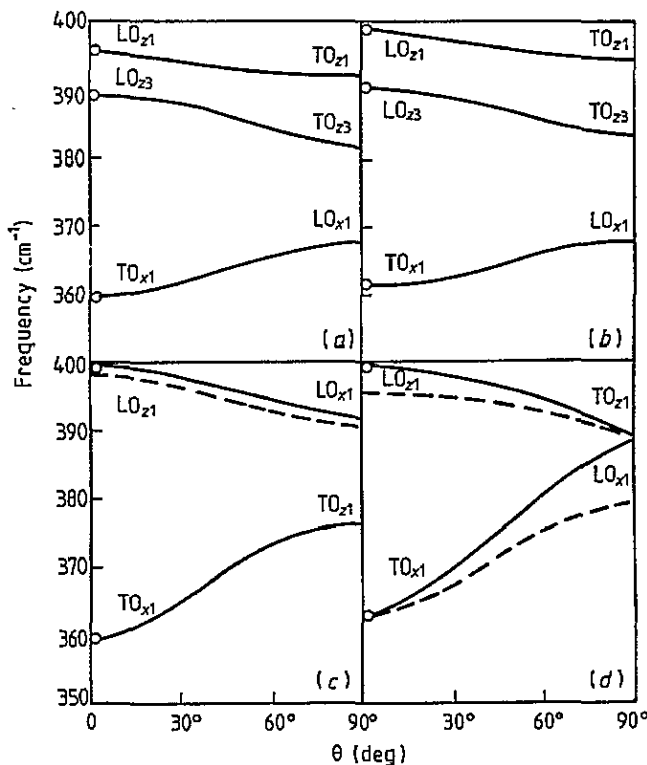

Figure 2. The angular dispersion of p-polarized optical vibrational modes in the sls (a) $(GaAs)_8/(AlAs)_3$, (b) $(GaAs)_{10}/(AlAs)_4$, (c) doped $(GaAs)_5/(AlAs)_5$ with $N = 1 \times 10^{17}$ cm^{-3}, and (d) doped $(GaAs)_5/(AlAs)_5$ with $N = 2 \times 10^{18}$ cm^{-3}: ○, values of frequencies of confined optical phonons measured by FTIR; – – – –, curves calculated with the same parameters but in the absence of free electrons.

Thus the different structural qualities of sls of the same type lead to different angular

dispersions of optical vibrational modes. For instance, the angular dispersion of optical modes obtained for two doped SLS with the same layer thicknesses but different doping levels is shown in figures 2(c) and 2(d). Apparently, the different doping levels cause different qualities of the samples. Only the first fundamental confined optical modes LO_{z1} and TO_{x1} have been observed in these samples. Therefore the angular dispersions of optical modes in these SLS have been calculated on the assumption that the high-index confined modes are damped by phonon scattering. The dashed curves in figures 2(c) and 2(d) show the angular variation in optical modes calculated with the frequencies of the confined modes and their oscillator strengths obtained from experimental reflection spectra but assuming the absence of free electrons.

Thus the influence of doping on the angular dispersion of vibrational modes in doped SLS is clearly seen. As a result of the plasmon–LO-phonon coupling the frequencies of longitudinal LO_x and LO_z modes are shifted to the high-frequency region.

We emphasize the fact that the frequencies of confined LO_z and TO_x modes were approximately the same in both samples but the frequencies of LO_x and TO_z modes were different as a result of the different doping levels and structural qualities of the samples.

In the case of one sample studied here (the angular dispersion of this sample is plotted in figure 2(c)) the frequency of the LO_x mode was higher than the frequency of the TO_z mode. This is the consequence of the high values of oscillator strengths $R_{tl}^{(2)}$ and $R_{ll}^{(2)}$ observed in this sample (see table 1). Figure 3 presents the calculated dependences of the frequencies of all vibrational modes observed in this sample on the oscillator strength $R_{tl}^{(2)}$ at $R_{ll}^{(2)} = 50\,000$ cm^{-2}. As is seen, the high values of oscillator strengths lead to the crossing of the LO_{x1} and TO_{z1} modes. The open circles in figure 3 display the values of mode frequencies obtained in the experiment.
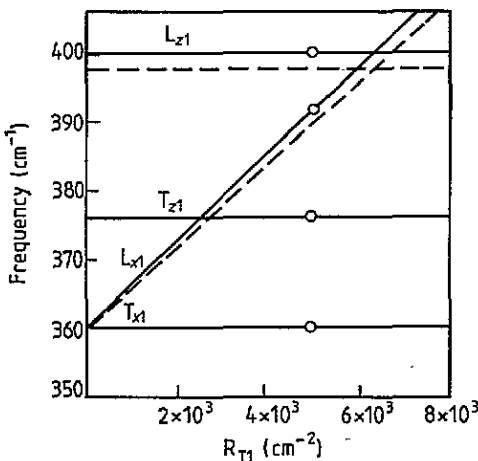

Figure 3. The calculated dependence of frequencies of optical vibrational modes observed for the doped $(GaAs)_5/(AlAs)_5$ SL with $N = 1 \times 10^{17}$ cm^{-3} on the value of oscillator strength $R_{tl}^{(2)}$ with $R_{ll}^{(2)} = 50\,000$ cm^{-2}: O, values of mode frequencies obtained in experiment; ----, curves with the same parameters but in the absence of free electrons.

In conclusion, in this paper we have shown that the analysis of p-polarized reflection spectra of SLS makes it possible to obtain values of the frequencies of optical modes propagating normal to the layers (confined modes) and parallel to the layers (interface

modes). As a result of infrared investigation of the anisotropy of zone-centre optical photons in $(GaAs)_n/(AlAs)_m$ SLs we can conclude that the angular dispersion of optical vibrational modes is an individual characteristic of each sample which strongly depends on its structural quality. In addition we observed that in doped SLs the coupling of longitudinal vibration modes with plasmons with corresponding electric polarization changes the angular dispersion drastically.

We hope that the results presented in this paper will be useful in order to characterize the quality of semiconductor SLs.

Acknowledgment

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