Light scattering by optical phonons trapped in AlAs layer in $(GaAs)_m(AlAs)_n$ superlattice

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(Submitted 5 June 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 1, 32–35 (10 July 1989)

A Raman scattering by *LO* phonons trapped within an AlAs layer in a $(GaAs)_m(AlAs)_n$ ($m = 7, n = 18$) superlattice has been observed. When specific atoms (Ga, Al, As) were put in Wyckoff positions, it was found that the contributions of the displacements of these atoms to vibrations of a certain symmetry Γ_1, Γ_2, Γ_5 depends on m and n. It thus becomes possible to extract information about the microstructure and the quality of the superlattices within a single monolayer.

In semiconductor superlattices consisting of alternating layers of two isostructural materials with thicknesses d_1 and d_2, in which the period of the superlattice satisfies $d = d_1 + d_2 < \lambda$ (λ is the mean free path of the carriers), new features (not

 0021-3640/89/130037-05$01.00

possible in the original materials) may be manifested because of quantum size effects for optical phonons, as in the case of electrons. For example, optical phonons which are propagating along the quantization direction (the growth direction) of a superlattice may turn out to be trapped. In $(GaAs)_m(AlAs)_n$ superlattices, where m and n are the numbers of GaAs and AlAs monolayers, respectively, in the unit cell (the thickness of one monolayer is 2.83 Å), the Raman scattering involving LO_l phonons trapped in the GaAs layer has been studied in most detail.[1-10] It is difficult to experimentally observe LO_l phonons (l is the trapping order) trapped in an AlAs layer during excitation of Raman spectra under conditions close to a resonance with exciton transitions between quantum-size levels because of a manifestation of spatially extended (surface) vibrations caused by the interface of the heterolayers.[11] Futhermore, structural disorder in the transition layer may cause vibrations not manifested in the spectra of high-quality crystals to become active in the Raman scattering. This circumstance imposes stiff requirements of the structural quality of the superlattice.

In this letter we are reporting the observation of a Raman scattering by LO_l phonons trapped in AlAs layer in a $(GaAs)_7(AlAs)_{18}$ superlattice with a transition-layer thickness of 3.64 Å and with layer thicknesses $d = 20.75$ Å (GaAs) and $d_2 = 51.45$ Å (AlAs) during excitation of the spectra far from resonances with exciton transitions.

The single-crystal superlattice samples were grown by molecular-beam epitaxy on n-GaAs substrates oriented along the [001] axis. The period of the superlattice was repeated 100 times.

Figure 1 shows some typical Raman spectra in the $(GaAs)_7(AlAs)_{18}$ superlattices during excitation by the beam from an argon laser with a wavelength of 5145 Å. These spectra were recorded at $T = 10$ K in the frequency region of optical phonons of AlAs and in the scattering configurations $z(x'x')\bar{z}$ and $z(x'y')\bar{z}$. A characteristic feature of these spectra is the presence of well-expressed auxiliary lines of LO_l phonons, which are a consequence of a trapping of phonons by the longitudinal optical branch of AlAs. The arrow marks the position of the LO_0 phonon line in the original unperturbed AlAs, with $\omega_{LO_0} = 405.8$ cm^{-1} at $\mathbf{k} = 0$. The measured frequencies of the LO_l phonons trapped in the AlAs layer for $l = 1$, 2, 3, and 4 are 405.0, 402.0, 397.5, and 396.0 cm^{-1}, respectively. These values agree well with theoretical data recently generated for a GaAs-AlAs superlattice on the basis of a rigid-ion model, with a short-range interaction between nearest and second-nearest neighbors and with a long-range interaction between ions in nearest layers.[12]

A method of band representations of space groups has been developed for analyzing the symmetry of phonons in superlattices.[13] For complex crystalline systems with a large number of atoms in the unit cell, that method is considerably more effective than the conventional approaches, since the procedure for constructing the band representations does not require information about the crystal structure. (Simple band reprelumns give the indices of the band representations in the $\mathbf{k}$ basis found by induction from those irreducible representations of the local group under which the components x, y, z transform. In other words, the last four columns show the indices of the irreducible representations of the group of the wave vector (in the notation of Ref. 14) which are unambiguously related to the irreducible representations of space group

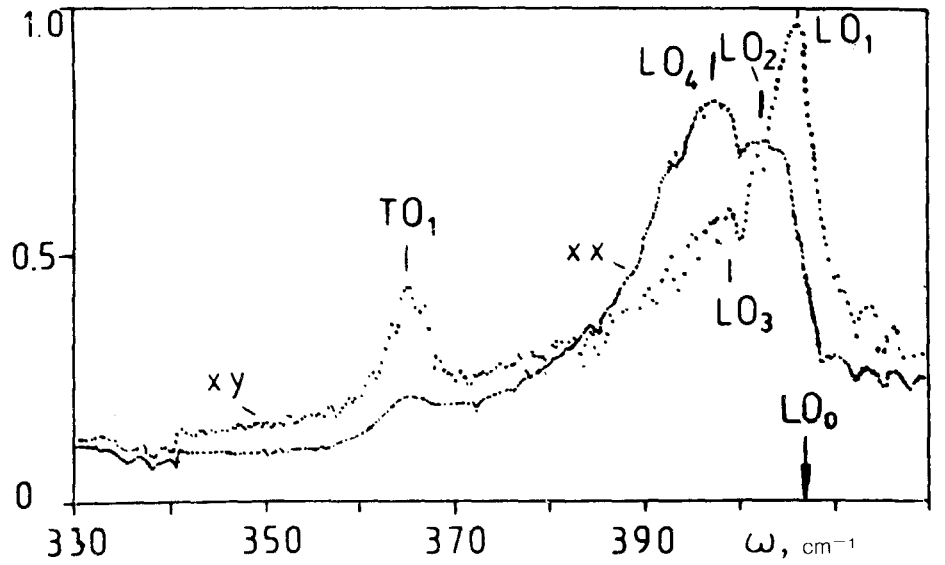

FIG. 1. Spectra of the scattering of light by LO_l phonons trapped in an AlAs layer in a $(GaAs)_7(AlAs)_{18}$ superlattice for the cases in which the polarizations of the incident and scattering light are crossed, $z(x'y')\bar{z}$ (phonons of Γ_2 symmetry with odd l are active), and parallel, $z(x'x')\bar{z}$ (Γ_1, with even l). The x', y', and z axes run parallel to the [100], [010], and [001] directions, respectively, and are rotated 45° around the z axis with respect to the translation vectors in the plane of the layer. $T = 10$ K; $\hbar\omega_i = 2.409$ eV.

sentations are constructed not for a specific crystal structure but for space group F.) We have established that the band-representation method is particullarly effective for analyzing the phonons symmetry in superlattices, in our case, superlattices based on III–V materials, whose symmetry is described by only two space groups, depending on the number of monolayers $(m+n)$ in the unit cell: $D^5_{2d}(m+n=2k)$ and $D^9_{2d}(m+n=2k+1)$. There are eight nonequivalent arrangements of atoms among symmetry positions (Wyckoff positions) for arbitrary m and n. By constructing band representations for the D^5_{2d} and D^9_{2d} groups and then placing atoms (Ga, Al, As) in the unit cell in the corresponding Wyckoff positions (we have derived general formulas for each of the eight types of atomic arrangements), we found the symmetry of the phonons for each of the sublattices having these symmetry groups. It follows from our analysis that for the $(GaAs)_7(AlAs)_{18}$ superlattice, whose symmetry is described by space group D^9_{2d}, the distribution of atoms among Wyckoff positions takes the following form: There is one Ga atom in site $1a$, with coordinates (000); there is one As atom in site $1c(0\frac{1}{2}\frac{1}{4})$; six Ga and 18 Al atoms are in pairs in sites $2e(00z)(00\bar{z})$; and 24 As atoms are in pairs in $2f(0\frac{1}{2}z)(\frac{1}{2}0\bar{z})$, positions, where the coordinates of the symmetry positions are expressed in units of the translation vectors of the tetragonal unit cell, and the symmetry of the phonons at the symmetry points of the Brillouin zone is as shown in Table I. The first column of this table gives the Wyckoff positions, along with the occupation of these positions by atoms; the second column shows the components of the vectors of local atomic displacements; and the third through sixth co-

TABLE I. Symmetry of the phonons in the $(GaAs)_7(AlAs)_{18}$ superlattice.

D^9_{2d}		Γ	M	X, P	N
1 Ga*(a)*	z	2	2	2	1
	x, y	5	5	3,4	1,2
1 As*(c)*	z	2	1	4	1
	x, y	5	5	1,2	1,2
3 × 2Ga*(e)*	z	1,2	1,2	1,2	1,1
9 × 2Al *(e)*	x	5	5	3,4	1,2
	y	5	5	3,4	1,2
12 × 2As*(f)*	z	1,2	1,2	3,4	1,1
	x	5	5	1,2	1,2
	y	5	5	1,2	1,2

D^9_{2d} and which determine the symmetry of the phonons at the corresponding symmetry points of the Brillouin zone.

From this table we can determine the contributions of the vibrations of specific atoms (Ga, Al, As) at certain Wyckoff positions to the phonon states. For example, the displacements of the following atoms contribute to the phonons with symmetry Γ_1 and Γ_2 observed experimentally: $24\Gamma_1(Ga^z_e; Al^z_e; As^z_f)$, $26\Gamma_2(Ga^z_{a,e}; Al^z_e; As^z_{c,f})$. It is important to note that these contributions, particularly for ultrathin superlattices, depend strongly on m and n, since a change in the number of layers is accompanied by a corresponding change in the distribution of atoms among Wyckoff positions. The structure of the vibrational representation also changes in the process; i.e., the phonon spectrum undergoes a substantial restructuring.

In summary, this new approach makes it possible to extract useful information about the crystal microstructure and the structural quality of a superlattice within a single monolayer from an analysis of Raman spectra.

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Translated by Dave Parsons