Multiphonon resonant Raman scattering and effects of tunneling of electronic excitations in short-period GaAs–AlAs superlattices

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The spectra of resonant Raman scattering of short-period GaAs–AlAs superlattices have been studied experimentally. Resonant Raman scattering processes of up to fourth order have been detected for the first time. The tunneling of electronic excitation in such systems is taken into account in an analysis of the results.

The optical properties of superlattices differ substantially from the characteristics of solid solutions of equivalent chemical composition. The reason lies in the spatial periodicity of superlattices along the direction perpendicular to the plane of the layers. Quantization gives rise to a new system of allowed phonon and electronic states, which is determined by the geometric thicknesses of the layers. Most of the existing data on vibrational excitations of superlattices have been obtained through Raman spectroscopy. The electronic structure of multilayer systems has been studied by the methods of photoreflection, photoluminescence, etc. In this letter we are reporting an experimental study of GaAs–AlAs superlattices by the method of resonant Raman spectroscopy.

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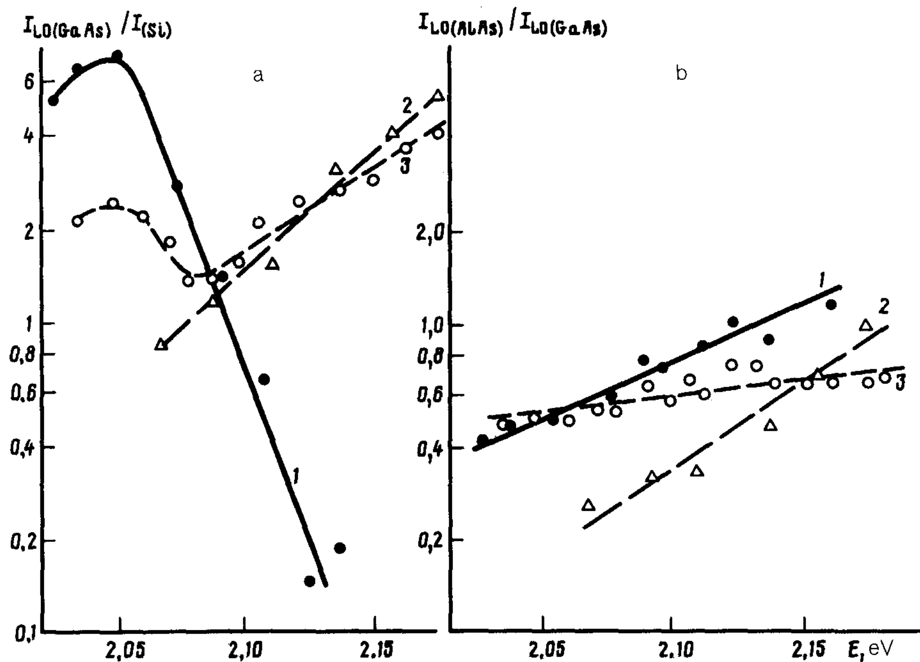

FIG. 1. a—Frequency dependence of the intensity of the light scattering by LO phonons of GaAs layers; b—frequency dependence of the relative intensities $I_{LO\,(\mathrm{AlAs})}/I_{LO\,(\mathrm{GaAs})}$ in superlattices with (1) $d_1 = d_2 = 2$ nm, (2) $d_1 = d_2 = 3$ nm, and (3) $d_1 = 1$ nm, $d_2 = 3$ nm.

This method provides information about both the vibrational modes of the structure, on the one hand, and the electronic states and particular features of the electron-phonon interaction, on the other.[1-3]

The GaAs–AlAs superlattices, not deliberately doped, were fabricated by molecular beam epitaxy on single-crystal GaAs substrates. The spectra were recorded on an apparatus based on a DFS-24 spectrometer with a photon counting system. The excitation source was a dye laser tunable over the energy interval 2.03–2.18 eV. All the measurements were carried out in a reflection geometry, at a sample temperature of 85 K. The spectral width of the slits did not exceed 3.0 cm^{-1}.

A characteristic feature of Raman scattering in superlattices is a resonant intensification of the scattering as the energy of the exciting photons approaches values corresponding to electronic (or excitonic) transitions from upper quantum levels of the light holes (LH) and heavy holes (HH) of the valence band to the lowest-lying quantum levels (C_n, $n = 1,2,...$) of the conduction band. Figure 1(a) shows data for three superlattices, with periods d_1 (GaAs) $= 2$ nm, d_2(AlAs) $= 2$ nm; $d_1 = d_2 = 3$ nm; and $d_1 = 1$ nm, $d_2 = 3$ nm [in all cases, the scattering intensity was normalized to the intensity of the Raman line at $\nu = 521\ \mathrm{cm}^{-1}$ (0.064 eV) of crystalline silicon]. For the first superlattice, the energy of the $H1$–$C1$ transition to the lowest-lying level in the conduction band is 2.05 eV, according to a calculation from the Kronig–Penney

model, and lies in the interval of excitation energies which was studied. Correspondingly, the plot of the scattering intensity versus the energy of the exciting photon has a clearly defined maximum, which shrinks markedly, by nearly two orders of magnitude, as the frequency is increased. For the two other samples, the change in the scattering intensity as a function of the frequency is slight. The reason for these results is that for the superlattice with $d_1 = 1$ nm, $d_2 = 3$ nm the energy of the lowest-energy transition, $H1$–$C1$, is 2.49 eV, so that the excitation occurs in the preresonance region in this case. For the sample with $d_1 = d_2 = 3$ nm, the $H1$–$C1$, $L1$–$C1$, and $H2$–$C1$ energies are 1.86, 1.95, and 2.02 eV, respectively. A luminescence of these short-period sublattices is observed in the longer-wavelength part of the spectrum, as a result of indirect X-Γ transitions.[4]

Although actual electronic (or excitonic) transitions occur only in the GaAs layers in these superlattices, as a result of the absorption of laser photons, the spectra of the scattering intensity are also of a resonant nature for the LO phonons of the AlAs component [Fig. 1(b)]. Furthermore, with increasing frequency of the exciting light the intensity of the LO(AlAs) line in fact increases in comparison with LO(GaAs). This effect is a consequence of a tunneling of electronic (or excitonic) excitations into the (AlAs) barrier region and is apparently a general effect in short-period superlattices. For superlattices with thick layers ($d \approx 20$ nm), a resonant intensification of light scattering in the barrier layers of this type is not seen.[5]

As was shown in Ref. 6, by taking the tunneling into account on the basis of a simple model one can find a qualitatively correct description of the frequency dependence of the relative intensities of the scattering lines of the GaAs and AlAs components.[2)] The approximately exponential increase in the relative intensity of the LO(AlAs) line in comparison with LO(GaAs) with increasing frequency occurs because the energy of the exciting photons, E_{las}, approaches the value corresponding to the band gap of the barrier layers, E_g^x(AlAs), so there is an increase in the barrier transmission:

$$\frac{I_{LO(\text{AlAs})}}{I_{LO(\text{GaAs})}} \sim \exp\{-[E_g^x(\text{AlAs}) - E_{\text{las}}]/E_B\}, \tag{1}$$

The parameter E_B is determined by the geometric thickness of the barrier, $d_B = d_2$: $E_B \sim 1/\sqrt{d_B}$ [expression (1) was derived in Ref. 6 under the assumption that the electron-phonon coupling constants in the well material and the barrier material are the same].

Figure 1(b) shows that as we go from the superlattice with $d_1 = d_2 = 2$ nm to that with $d_1 = d_2 = 3$ nm, we observe (on the one hand) a decrease in this ratio of line intensities at a fixed excitation frequency and (on the other) an increase in the slope of the frequency dependence of this ratio. These facts can be described in a qualitatively correct fashion on the basis of the simple model which we mentioned above. On the other hand, a decrease in the thickness of the quantum-well layers (GaAs) should not determine the frequency dependence of the intensity ratio of these scattering lines according to (1). While we see from Fig. 1(b) that the frequency dependence is quite different for curves *2* and *3*, which correspond to superlattices with an identical barrier thickness $d_2 = 3$ nm. This result is evidence that the model used here is oversimplified.

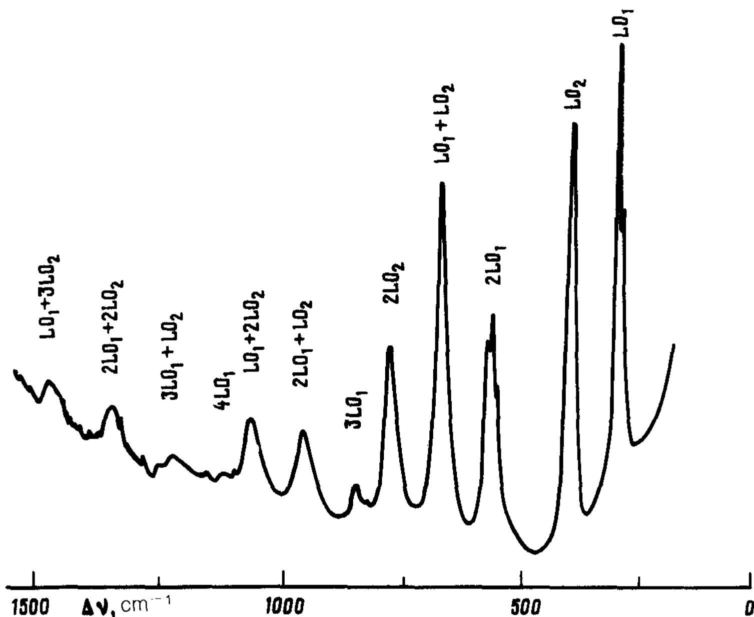

FIG. 2. Spectrum of the multiphonon resonant Raman scattering of a superlattice with $d_1 = d_2 = 2$ nm at $E_{\text{las}} = 2.163$ eV. Subscripts 1 and 2 correspond to the LO phonons of the GaAs and AlAs layers, respectively.

The absolute values and the ratio of the thicknesses of the layers of the short-period superlattice as well as the difference between the electron-phonon coupling constants of its components should evidently be taken into account. As we know, the difference substantially determines resonant Raman scattering in single crystals of semiconductors[7] and solid solutions based on them.[8]

That a tunneling of electronic excitations plays a governing role is supported by the resonant nature of the scattering of light by the LO phonons of the AlAs component, even in the case in which actual transitions occur only in the GaAs layer as a result of the absorption of laser photons. Consequently, for short-period superlattices an electron-hole excited by the light will, with a certain probability, be in both GaAs and AlAs layers. Furthermore, the thermalization of an electron-hole pair of this type may occur in a process involving excitation of LO phonons of both GaAs and AlAs, as can be seen from the spectra of the multiphonon resonant Raman scattering. It can be seen from Fig. 2 that not only the overtone lines nLO(GaAs) and nLO(AlAs) but also the composite lines lLO(GaAs) + KLO(AlAs) of up to fourth order are clearly present in these spectra.[3] As in the case of semiconducting solid solutions,[10] the relative intensities of the composite lines increase with increasing scattering order. For example, while the intensities of the first-order lines LO(GaAs) + LO(AlAs) are comparable, those of the composite lines LO(GaAs) + LO(AlAs) and $2LO$(GaAs) + LO(AlAs) are several times higher than those of the overtone lines of the same

order. An important point here is that the GaAs and AlAs components are spatially separate in the case of superlattices, in contrast with the case of a solid solution.

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[2] Only a single superlattice, with layer thicknesses $d_1 = 1.25$ nm and $d_2 = 3.75$ nm, was studied in Ref. 6.

[3] The fine structure in the LO(GaAs) and $2LO$(GaAs) scattering lines stems from the "capture" of optical phonons which is characteristic of size-effect superlattices.[9]

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