Raman scattering in GaAs/AlAs structures with paired quantum wells

V. A. Gaĭsler, D. A. Ténné, A. O. Govorov, N. T. Moshegov, A. I. Toropov, and A. P. Shebanin

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

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Both nonresonant and resonant Raman scattering of light by longitudinal optical phonons have been studied in GaAs/AlAs structures with paired quantum wells. Because of the particular shape of the envelopes of the electron wave functions and potentials set up by the phonons, it was found possible, for the first time, to observe a scattering by odd and even localized *LO* phonons simultaneously under nonresonant conditions. The absence of peaks corresponding to odd phonons from the Raman scattering spectrum is explained on the basis of an additional scattering mechanism: a Fröhlich interaction which is induced by defects and which leads to the predominance of even modes in the spectra.

Optical phonons localize in layers of a single material in GaAs/AlAs superlattices.[1–3] This effect is manifested in the Raman scattering spectra by several peaks, whose frequencies correspond to the frequencies of optical phonons of the bulk material, for wave vectors

$$q=\frac{m_\pi}{(n+1)a_0},\quad m=1,2,3..., \tag{1}$$

where a_0 is the thickness of GaAs monolayer, and n is the number of monolayers.[1,2] In GaAs/AlAs superlattices grown in the (001) direction, in the backscattering geometry $z(x,y)\bar{z}$ (the z axis runs perpendicular to the layers of the superlattice), only odd localized *LO* phonons ($m=1,3,5,...$) are manifested in the Raman spectra under nonresonance conditions. A scattering by even phonons is observed in the parallel geometry $z(x,x)\bar{z}$ under resonant conditions.[1–3] In this letter we are reporting the observation, under nonresonant conditions, of a scattering by odd and even localized *LO* phonons simultaneously in a GaAs/AlAs superlattice with paired quantum wells.

We studied both nonresonant and resonant Raman scattering in samples of a structure consisting of 40 $(GaAs)_{12}(AlAs)_3(GaAs)_{12}(AlAs)_{12}$ periods. Each period constituted two GaAs quantum wells separated by a thin AlAs barrier. We also studied Raman scattering in an ordinary $(GaAs)_{12}(AlAs)_{12}$ superlattice (these were samples *A* and *B*, respectively). The samples of both types were grown by molecular beam epitaxy on GaAs (001) substrates on apparatus developed at the Institute of Semiconductor Physics, Novosibirsk. The layer thicknesses were monitored during the growth by measuring the oscillations in the intensity of the specular reflection in reflection high-energy electron diffraction. The Raman spectra under nonresonant conditions were measured during excitation by the beam from an argon laser with a

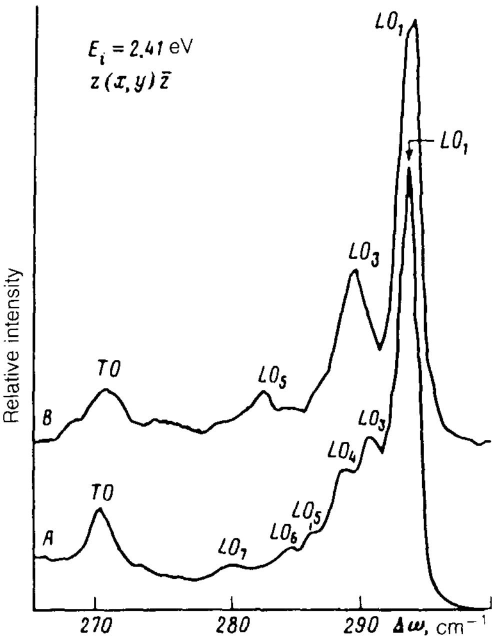

FIG. 1. Spectra of nonresonant Raman scattering of samples *A* and *B*.

wavelength of 514.5 nm at a temperature of 77 K in a quasi-back-scattering geometry $z(x,y)\bar{z}$. To observe the resonant Raman scattering we used a tunable Ti:Al_2O_3 laser pumped by an argon laser. The spectra of resonant Raman scattering were measured in the parallel geometry $z(x,x)\bar{z}$.

Figure 1 shows the Raman spectra of samples *A* and *B* under nonresonant conditions, and Fig. 2 shows the corresponding spectra under resonant conditions. The spectra of sample *B* correspond to the selection rules stated above: In the geometry $z(x,y)\bar{z}$, under nonresonant conditions, we find odd localized phonons LO_1, LO_3, and LO_5. In the parallel geometry, under resonant conditions, we find the even phonons LO_2, LO_4, and LO_6; we also find some interfacial (IF) phonons. The spectrum of resonant Raman scattering in sample *A* also has some even and interfacial phonons, but the spectrum of the nonresonant scattering has peaks corresponding to both odd and even localized *LO* phonons. (The frequency difference between the LO_1 and LO_2 phonons is only 0.2 cm^{-1}, so they could not be resolved.)

We offer the following explanation for the appearance of peaks corresponding to both odd and even localized *LO* phonons in the spectrum of nonresonant Raman scattering of sample *A*. The intensity of Raman scattering for a process involving the emission of a single phonon is proportional to[4]

$$W_{fi}=\sum_{a,\beta}\frac{\langle f|\hat{H}_{E-R}|\alpha\rangle\langle\alpha|\hat{H}_{E-P}|\beta\rangle\langle\beta|\hat{H}_{E-R}|i\rangle}{(\hbar\omega_s-E_\beta+i\Gamma_\alpha)(\hbar\omega_i-E_\alpha+i\Gamma_\beta)}. \tag{2}$$

Here ω_i and ω_s are the frequencies of the incident and scattered light. The scattering from the initial state ($|i\rangle$) to the final state ($|f\rangle$) occurs through intermediate

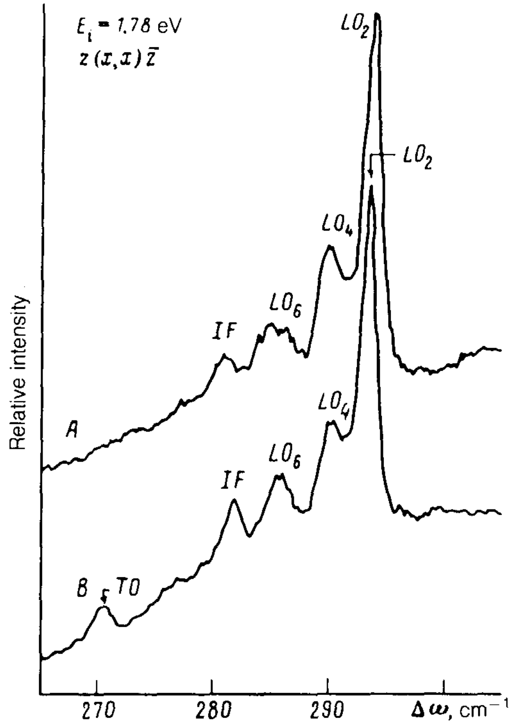

FIG. 2. Spectra of resonant Raman scattering of samples *A* and *B*.

electron–hole-pair state $|\alpha\rangle$ and $|\beta\rangle$, which have energies $E_{\alpha,\beta}$ and widths $\Gamma_{\alpha,\beta}$. The quantities $\hat{H}_{E-R}$ and $\hat{H}_{E-P}$ are the Hamiltonians of the interaction of electrons and holes with electromagnetic radiation and phonons, respectively. The electron–phonon Hamiltonian can be written $\hat{H}_{E-P}=\hat{H}_{DP}+\hat{H}_F$, where $\hat{H}_{DP}$ is the interaction through the strain energy, and $\hat{H}_F$ is the Fröhlich interaction, which is associated with macroscopic electric fields set up by *LO* phonons.[3,4]

The electron (hole) wave function $\Phi_{e(h)}$ can be written as the product of a rapidly varying Bloch function $u_{e(h)}$, a slowly varying envelope $\Psi n_{e(h)}$, which describes the localization of electrons (holes) along the z direction in the $n_{e(h)}$ subband, and a plane wave which describes the free motion in the direction parallel to the plane of the quantum well (the coordinates x,y):

$$\Phi_{e(h)} \sim e^{i\mathbf{k}_{e(h)}\mathbf{r}_{xy}} u_{e(h)}(\mathbf{r})\Psi n_{e(h)}(z). \qquad (3)$$

The selection rules listed above follow from the symmetry of the envelopes of the electron wave functions $\Psi n_{e(h)}(z)$ and of the potentials set up by the phonons (strain or Fröhlich potentials) with respect to the plane passing through the center of a well. In sample *A* there are two GaAs quantum wells, separated by an AlAs barrier 3 monolayers thick. For phonons, this barrier is fairly thick (the *LO* phonon of GaAs penetrates a depth ~ 1 monolayer into the AlAs layer), and the displacements of the atoms and thus the potentials set up by the phonons are essentially the same as for a single ionized well. The symmetry with respect to the center of each of the wells separately is preserved. The electrons, in contrast, can easily penetrate through a

3-monolayer barrier, and the envelopes of the electron wave functions have the symmetry of the overall system, i.e., a symmetry with respect to a plane passing through the center of the thin AlAs barrier. The selection rules are thus violated, and both even and odd localized *LO* phonons should be seen in the Raman spectra under both nonresonant and resonant conditions.

We believe that the absence of the peaks corresponds to odd modes from the spectrum of sample *A* under resonant conditions can be explained in the following way: A more complex scattering mechanism, which involves (in addition to an electron–phonon interaction) an elastic scattering of an electron by impurities, defects, or disordered heterojunctions (a Fröhlich interaction induced by defects), is operating.[4,5] A phonon involved in a scattering may have a nonzero wave-vector component q_{xy}, parallel to the layers of the quantum well. This process of double scattering under resonant conditions may outweigh the process described by Eq. (2) (Ref. 5). Support for the idea that this mechanism is operating comes from the presence of peaks corresponding to interfacial phonons, which also have a nonzero component q_{xy}, in the spectra of the resonant Raman scattering of both samples.

If a phonon has a wave-vector component parallel to the layers of the quantum well, it may have a substantial influence on the intensity of scattering by localized *LO* phonon with various indices m. The frequencies of odd phonons differ from those of even phonons in that they depend on the direction of the wave vector even at values[6] $q \to 0$. The peaks corresponding to odd modes may thus be shifted in the low-frequency direction and substantially broadened. These effects may explain why we do not observe these peaks in the resonant Raman-scattering spectrum of sample *A*.

In summary, we have reported the first observation of a Raman scattering by odd and even localized *LO* phonons simultaneously in a GaAs/AlAs structure with paired quantum wells under nonresonant conditions. The selection rules are violated because of the particular symmetry of the envelopes of the electron wave functions and the potential set up by the phonons. We do not, on the other hand, observe the appearance of odd phonons in the spectrum of resonant Raman scattering. We attribute this result to the operation of an additional scattering mechanism: a Fröhlich interaction induced by defects, which involves phonons with a nonzero wave-vector component parallel to the structural layers. The manifestation of this mechanism under resonant conditions has the consequence that even localized *LO* phonons are predominant in the Raman spectra.

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