Resonant Raman scattering and redislocation effects in GaAs/AlGaAs

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(Submitted 30 December 1993)
Pis'ma Zh. Eksp. Teor. Fiz. **59**, No. 4, 235–239 (25 February 1994)

A redislocation of electronic excitations out of a barrier into quantum wells of a GaAs/AlGaAs quantum-well structure upon a narrowing of the barriers is predicted theoretically. This effect has also been seen experimentally, by the method of resonant Raman spectroscopy. The possibilities of resonant Raman spectroscopy for testing the localization of the electron wave function in quantum-well structures are demonstrated.

The redislocation effect, i.e., a change in the position of the maximum of an electron wave function and thus a redistribution of electrons among nonequivalent quantum wells of a quasi-2D structure upon the application of an external agent, underlies the operation of a new class of nanoelectronic devices.[1] Studies have been made of the structure of the subbands and their populations for nonequivalent quantum wells[2] and of the influence of the quantum-size effect on the frequencies of optical phonons of isolated quantum wells.[3] These studies have been carried out by the method of Raman scattering. Encouraging results have been found. They raise the hope that resonant Raman spectroscopy will prove effective for probing the redislocation effect. In this letter we report, as a first step toward the solution of this problem, a study of redislocation from barriers into quantum wells in GaAs/$Al_xGa_{1-x}As$ superlattices by the method of resonant Raman spectroscopy.

GaAs/$Al_xGa_{1-x}As$ superlattices consisting of 40 periods of quantum wells 3 nm thick and of barrier layers with a nominal composition $x=0.35$, whose thickness d_b ranged from 20 to 2 nm in different samples, were grown by molecular beam epitaxy on (100)GaAs substrates. By varying the thickness of the barrier layers we were able to vary the degree of confinement of the electron states in the quantum-well and barrier layers and also the structure of the minibands. The composition of the barrier layers was monitored within 0.01 on the basis of the difference between the frequencies of AlAs-like and GaAs-like LO modes of $Al_xGa_{1-x}As$ in the Raman scattering spectra.[4] This composition turned out to be approximately $x=0.34$. Raman spectra were excited by the beam from a laser using a mixture of Ar and Kr or of He and Ne, with discrete wavelengths spanning the range 488–676.4 nm (2.54–1.83 eV). The Raman spectra were analyzed with a Ramanor U1000 spectrometer with a resolution of 1–3 cm^{-1}. These spectra were recorded in a single-channel arrangement with a calibration based on lines from the discharge of a Ne lamp. The measurements were carried out at a temperature of 300 K.

The photoluminescence spectra of a structure with $d_b=20$ nm, measured at 300 K, contain an intense band peaking at 1.56 eV, which is due to transitions from the

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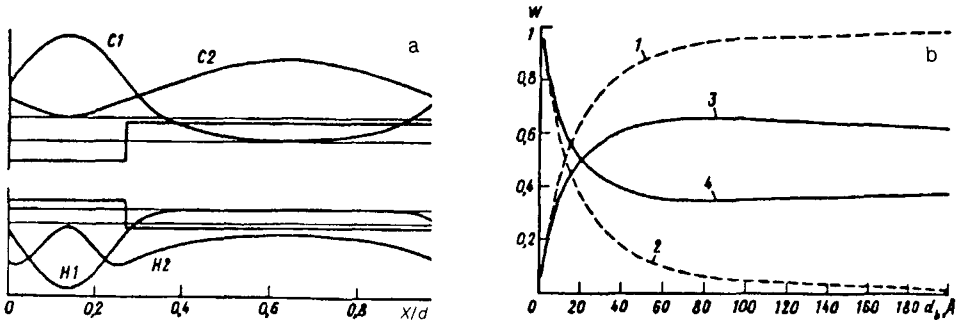

FIG. 1. a: Energy-level diagram and profile of the wave functions $[|\Psi(X)|^2]$ along the growth axis, X (X is divided by the period of the structure, d), for the first and second subbands of the conduction band ($C1$ and $C2$) and for the heavy-hole subbands ($H1$ and $H2$) in a structure with $d_b=8$ nm. b: Relative probabilities for finding an electron (W_e) and a hole (W_h) in the barriers and quantum wells of a quantum-well structure versus d_b. *1, 2*—W_e for barriers and wells, respectively; *3, 4*—W_h for barriers and wells.

lower subband of the conduction band ($C1$) into upper subbands of light and heavy holes[5] ($L1$ and $H1$). In addition, we detected a weak band at 1.88 eV. On the basis of calculations of the miniband structure by the envelope method and an estimate of the matrix elements of transitions of approximately the same energy, this band was assigned to a transition between the second conduction subband ($C2$) and the second heavy-hole subband ($H2$). As the barrier is narrowed, this peak shifts up the energy scale, and its position correlates well with a theoretical prediction of the energy of the $C2$–$H2$ transition. Figure 1a shows a theoretical schematic diagram of the energy levels and a plot of the profile of the wave functions $[|\Psi(X)|^2]$ along the growth axis, X (X is divided by the period of the structure, d). The results here correspond to the first and second subbands of the conduction band ($C1$ and $C2$) and the heavy-hole subbands ($H1$ and $H2$) in a structure with $d_b=8$ nm. We see that the $C2$ subband lies above the barrier, while the $H2$ subband is in a well. The wave functions of the states corresponding to the $C2$–$H2$ transition are localized to a greater extent in the AlGaAs barriers. Our calculations show that this picture is also typical of the structure with $d_b=20$ nm. The possible localization of the electron wave function in the barriers of an $Al_xGa_{1-x}As/Al_yGa_{1-y}As$ superlattice was also predicted in Ref. 6 on the basis of an empirical strong-coupling model. A localization of this sort can explain transitions between states above the barrier in $GaAs/Al_xGa_{1-x}As$ superlattices, which have been observed previously by resonant Raman spectroscopy[7] and by photoluminescence excitation spectroscopy.[8] In the structures with $d_b=4$ and 2 nm, levels $C2$ and $H2$ are above the barrier. In addition, there is a significant probability for a localization of electrons in the quantum wells. This situation is demonstrated by Fig. 1b, which shows the d_b dependence of the relative probabilities for finding an electron (W_e) and a hole (W_h) in the barriers and in the wells of the quantum-well structure. (Curves *1* and *2* correspond to W_e for the barriers and the wells, while curves *3* and *4* correspond to W_h for the barriers and the wells). These curves were plotted by integrating $|\Psi(X)|^2$ along X. It follows from Fig. 1b that for structures with $d_b=20$ and 8 nm the

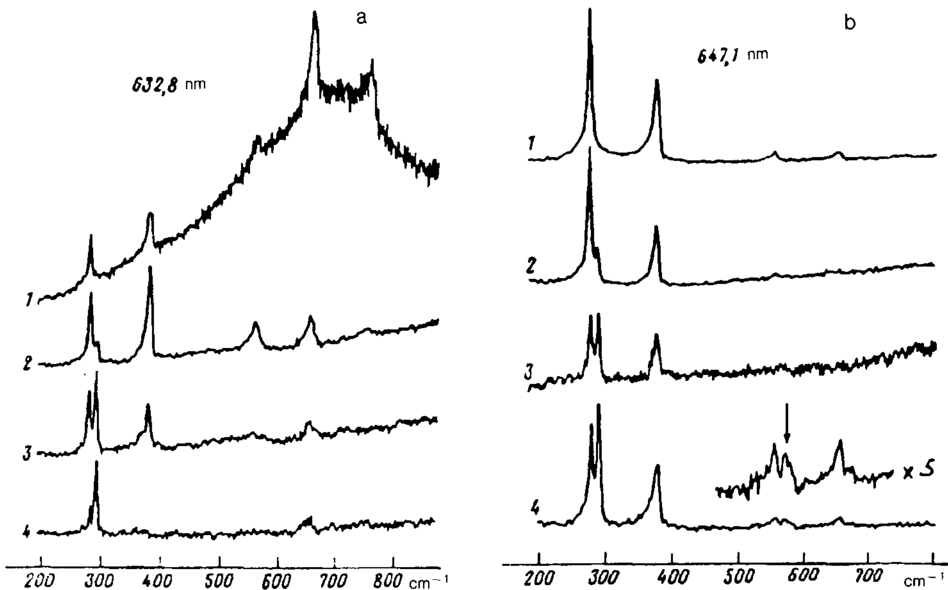

FIG. 2. a: Raman spectra of four GaAs/AlGaAs superlattices, with $d_b=20$, 8, 4, and 2 nm (spectra *1–4*, respectively), recorded at a temperature of 300 K, with excitation at the wavelength 632.8 nm. b: The same, but with excitation at a wavelength of 647.1 nm.

probabilities for finding an electron in the AlGaAs barriers are close to one (curve *1*), while at $d_b=4$ and 2 nm there is a significant probability for finding an electron in a well (curve *2*). For holes, on the other hand, the probabilities to be in a well and a barrier are approximately equal over the entire range $2\leqslant d_b\leqslant 20$ nm (curves *3* and *4*). Significantly, the $W(d_b)$ curves are very nonlinear; i.e., they cannot be explained in terms of a simple decrease in the relative weight of the barriers as these barriers become narrower.

Distinctive features of resonant Raman scattering by optical phonons as the photon energy of the exciting light, $E_{\rm ex}$, approaches the energy of a characteristic electronic transition are a multiple increase in the intensity of first-order modes and the appearance of overtone and composite lines in the spectra. The intensities of these lines far from resonance are less than that of the first-order Raman lines by a factor of hundreds. The higher-harmonic (overtone) and composite lines are amplified even further if $E_{\rm ex}$ is chosen in such a way that the photon energy of the scattered radiation is near this transition (the so-called outgoing resonance). These features are demonstrated clearly in Fig. 2a, which shows Raman spectra of four GaAs/AlGaAs superlattices, with $d_b=20$, 8, 4, and 2 nm (spectra *1–4*, respectively). These spectra were excited at a wavelength of 632.8 nm and recorded at a temperature of 300 K. Under these conditions, $E_{\rm ex}$ (1.96 eV) for the structure with $d_b=20$ nm is above the energy of the *C2–H2* transition by an amount roughly equal to the energy of an *LO* phonon. This transition falls in the region of $2LO$ scattering. In the spectra in Fig. 2, the peaks at 279 and 378 $\rm cm^{-1}$ stem from respectively GaAs-like and AlAs-like *LO* modes of

the AlGaAs barriers.[4] The peaks in the region of the $2LO$ scattering are assigned to the $2LO$ overtone of a GaAs-like mode (558 cm^{-1}), to a composite line $LO_{GaAs}+LO_{AlAs}$ (657 cm^{-1}), and to the $2LO$ overtone of the AlAs-like mode of AlGaAs (756 cm^{-1}). The energy of the $C2$–$H2$ transition for the structure with $d_b=8$ nm lies near 1.96 eV, so this transition shifts into the region of $1LO$ scattering (spectrum *2* in Fig. 2a). The same conditions are realized for the structure with $d_b=20$ nm during excitation by radiation with a wavelength of 647.1 nm ($E_{ex}=1.916$ eV) in Fig. 2b.

The intensity of the composite line in the Raman spectra in Fig. 2 is greater than the intensity of the overtone modes. This result is a consequence of a cascade process of resonant Raman scattering with the sequential production of each phonon in an individual electron–phonon interaction event.[9,10] In this process the rate of phonon production can be arbitrary. Each interchange results in an additional contribution to the scattering efficiency. As a result, the scattering efficiency for the composite lines has more contributions than that for the overtones, so the intensity of the composite lines is higher. Note also the high intensity of the AlAs-like LO mode in the spectra, despite the circumstance that the AlAs concentration in the AlGaAs barriers is 34%. We link this circumstance with a large value of the Fröhlich electron–phonon coupling constant for the AlAs-like LO mode in comparison with the GaAs-like mode. This interaction dominates the scattering efficiency for longitudinal vibrations near the resonance.

We wish to stress that the resonant Raman scattering is observed in this study near the $C2$–$H2$ transition, which is formed by states with a predominant distribution of wave functions in the barrier region (Fig. 1, a and b). We use specifically this circumstance to explain the resonant intensification of only the LO phonons of the AlGaAs barriers in the Raman spectra of superlattices with thick barriers ($d_b=20$ and 8 nm in Fig. 2). However, as the barrier becomes narrower, the relative intensity of the LO mode of the GaAs quantum wells increases near 292 cm^{-1}; this mode becomes predominant in the spectra of superlattices with $d_b=2$ nm (spectra *4* in Fig. 2, a and b). The effect observed here stems from a combination of many factors, among them the detuning from resonance, the values of the Fröhlich coupling constants, the relative probabilities for finding electrons and holes in the barriers and quantum wells, and the increase in the effective thickness of the probed layer of GaAs upon a narrowing of the barriers. However, the increase in the relative intensity of the LO mode of the GaAs quantum wells can be explained at a qualitative level in terms of a redislocation of electronic (excitonic) excitations out of the barrier region into the quantum wells, in accordance with Fig. 1b. The validity of our arguments is also supported by the onset of an additional structural feature near 570 cm^{-1} (marked by the arrow) in spectrum *4* in Fig. 2b. The intensity of this feature is comparable to that of the $LO_{GaAs}+LO_{AlAs}$ composite line of the AlGaAs barrier in the same spectrum. The position of this new line agrees well with the frequency of the composite mode of an LO phonon of a GaAs well and of a GaAs-like LO phonon of an AlGaAs barrier. The formation of such composite modes of spatially separated components of the superlattice becomes possible in structures with narrow barriers, for which the relative probabilities for an electron to be in the wells and the barriers are close in magnitude

(Fig. 1b). This circumstance is the fundamental distinction between the case under consideration here and the tunneling of electronic excitations through narrow barriers in the course of resonant Raman scattering near the $H1$–$C1$ transition between states localized in quantum wells.[9] In the latter case, the spectra of the resonant Raman scattering also contain some composite modes of spatially separated components of the superlattice.

In summary, this study has confirmed the theoretical prediction that electronic states may become localized in the barriers of GaAs/AlGaAs superlattices. A redislocation of electronic excitations from barriers into quantum wells upon a narrowing of the barriers has been observed. The possibilities of resonant Raman spectroscopy for testing the localization of an electron wave function in a quantum-well structure have been demonstrated.

This study had partial financial support from the Russian Basic Research Foundation (Project codes 93-02-2361 and 93-02-2362).

[1] A. A. Gorbatsevich *et al.*, Appl. Phys. Lett. (in press).
[2] D. Richards *et al.*, Appl. Phys. Lett. **56**, 1649 (1990).
[3] H. Tanino *et al.*, Appl. Phys. Lett. **60**, 1978 (1992).
[4] N. Saint-Cricq *et al.*, J. Appl. Phys. **61**, 1206 (1987).
[5] R. Cingolani and K. Ploog, Advances in Phys. **40**, 535 (1991).
[6] Lok C. Lew Yan Voon, *et al.*, Phys. Rev. B **47**, 6585 (1993).
[7] J. E. Zucker *et al.*, Phys. Rev. B **29**, 7065 (1984).
[8] J. J. Song *et al.*, Phys. Rev. B **34**, 8958 (1986).
[9] A. M. Brodin *et al.*, JETP Lett. **51**, 178 (1990).
[10] D. J. Mowbray *et al.*, Phys.Rev. B **43**, 11815 (1991).

Translated by D. Parsons