Differential spectroscopy of GaAs-$Ga_{1-x}Al_xAs$ quantum wells: An unambiguous identification of light-hole and heavy-hole states

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We present differential photoluminescence and reflectivity studies of GaAs-$Ga_{1-x}Al_xAs$ quantum wells, using both wavelength-modulation and piezomodulation techniques. Our experiments permit unambiguous identification of light- and heavy-hole exciton transitions.

For many years the optical properties of GaAs-$Ga_{1-x}Al_xAs$ quantum wells and superlattices structures have been extensively studied both experimentally and theoretically. The dominant optical techniques used have been absorption,[1,2] photoluminescence,[3-8] photoluminescence excitation,[7-9] and Raman spectroscopy.[10,11] Recently, low-temperature reflectance measurements[12] and room-temperature modulated reflectance measurements[13] have been reported. Low-temperature photoluminescence measurements have permitted observation of, essentially, the recombination lines associated respectively with the $n=1$ electron-heavy-hole (e-HH) $1s$ exciton, the $n=1$ electron-light-hole (e-LH) $1s$ exciton,[14,15] and more recently the $n=1$ (e-HH) $2s$ exciton state.[16] Absorption, reflectance, and excitation spectroscopy measurements, on the other hand, permit us to reveal many structures not apparent in photoluminescence studies, like, for instance, higher-lying interband optical transitions and structure splitting attributed to interfacial defects.

Now, all the experimental techniques quoted above distinguished between different energy levels without further information concerning the identity of the energy states. Particularly, it is not easy to experimentally identify the e-HH exciton states from the e-LH ones. In the present paper we report an unambiguous identification of these states from a comparative study of differential-spectroscopy spectra obtained respectively by the piezomodulation technique (PMT) and the wavelength-modulation technique (WMT). Concerning PMT it is worth noting that, following the relative magnitudes of the hydrostatic and shear deformation potentials of the GaAs valence band, the e-HH and e-LH exciton states are not modulated in a same way. In WMT, on the other hand, the modulation parameter is the same for both light- and heavy-hole exciton states. This is an essential difference between PMT and WMT concerning the differential spectroscopy of the exciton states. In other words, piezomodulation and wavelength modulation are, respectively, selective and nonselective differential spectroscopies.

We report low-temperature differential reflectivity and photoluminescence spectra in GaAs-$Ga_{1-x}Al_xAs$ multiple-quantum-well structures (MQW), with aluminum content $x=0.25$, well and barrier widths $L_z=60$ Å, and $L_b=70$ Å, respectively. The growth conditions of the samples have been described in preceding papers.[7]

Unmodulated photoluminescence (PL) and reflectivity (R) spectra, wavelength-modulated piezomodulated photoluminescence [(WPL), (PPL)] reflectivity [(WR), (PR)] spectra are taken at variable temperatures between 1.5 and 300 K. PL was excited with the 6328-Å line of a He-Ne laser, focused onto the entrance slit of a Jobin-Yvon 1.5-m focal-length grating monochromator and detected using a thermoelectrically cooled GaAs photocathode photomultiplier in the standard synchronous mode. In reflectance measurements the focused broadband light from a 100-W tungsten filament bulb was reflected from the sample onto the monochromator slit and subsequently dispersed and detected. In piezomodulation configuration the samples are glued onto a piezoelectric transducer excited by a low-frequency alternating electric field. Oscillating stress is applied in a coplanar configuration, so that the light is normally incident onto a (001) face, with the stress applied in the (001) plane. The wavelength-modulation spectra were recorded using a slight modulation of the output mirror of the monochromator.

Low-temperature reflectance (solid line) and photoluminescence (dotted line) spectra in the energy range of the MQW structures are given in Fig. 1. The reflectivity spectrum clearly shows two pairs of structures associated, respectively, to the CB_1-HH_1 (C_1H_1, C_1H_1') and CB_1-LH_1 (C_1L_1, C_1L_1') transitions. These doublets result from interfacial defects which are very large and at least greater than the exciton Bohr diameter (~ 300 Å) in order to trap the exciton and not to enlarge the exciton states. The interfaces consist of large growth islands differing by one monolayer height. It is worthwhile to note that the extra structure which appears on the low-energy side of the C_1H_1 structure is not a reflectance

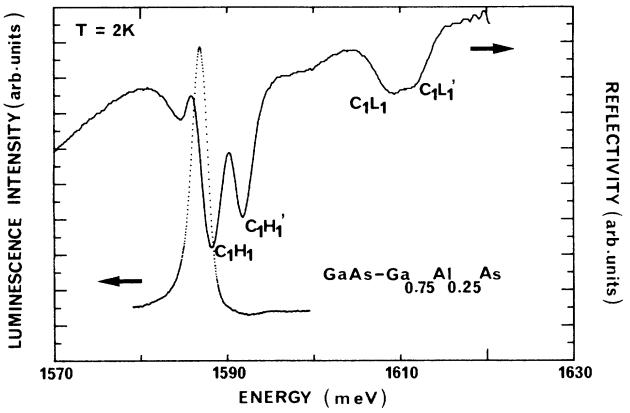

FIG. 1. Low-temperature reflectance (solid line) and photoluminescence (dotted line) spectra in the energy range of the MQW structure. Free exciton splitting due to well width fluctuations is evident.

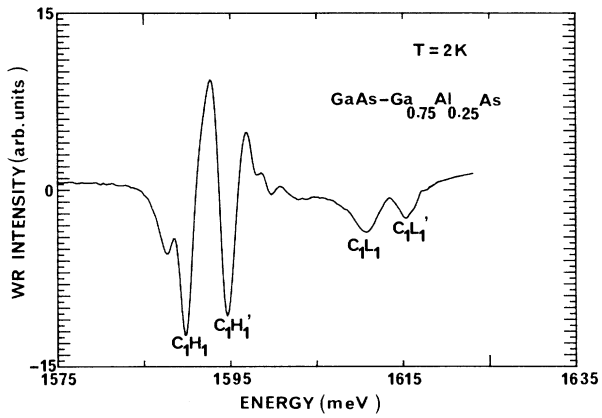

FIG. 2. Wavelength-modulated reflectivity spectrum.

one but results from the luminescence emission superimposed on the reflectance signal. This has been verified by exciting the sample with different laser intensities during the reflectivity measurements. Now let us compare the PL and reflectivity spectra; clearly the luminescence line appears on the low-energy side of the reflectivity structure. On the other hand, a theoretical analysis of the reflectivity spectrum in terms of exciton polariton shows that the reflectivity minimum does not correspond with the resonance energy E_T of the transverse exciton[17] but more precisely with the energy E_L of the longitudinal exciton. The energy difference $E_L - E_T$ which is a result of the long-range contribution of the electron-hole exchange energy is proportional to the oscillator strength of the exciton and may be important in two-dimensional (2D) structures. The exciton-polariton theory[17] shows that the resonance energy of the transverse exciton corresponds to the low-energy side of the reflectivity structure. In a first approximation, and to avoid tedious calculations, we may associate the resonance energy of the exciton with the inflection point which appears on the low-energy side of the reflectivity structure. This agrees with the energy of the luminescence-line maximum which appears on the low-energy side of the reflectivity structure.

The wavelength-modulated reflectivity spectrum is displayed in Fig. 2. In agreement with the preceding discussion, the exciton energy corresponds to the low-energy-side inflection point of the reflectivity spectrum and then to the minimum of the differential reflectivity spectrum. We measure $E(C_1H_1)=1589.5\pm0.2$ meV, $E(C_1H_1')=1594.3\pm0.2$ meV, $E(C_1L_1)=1610\pm0.3$ meV, $E(C_1L_1')=1614.5\pm0.3$ meV, and then $E(C_1H_1') - E(C_1H_1) = 4.8\pm0.4$ meV and $E(C_1L_1') - E(C_1L_1) = 4.5\pm0.6$ meV.

We have calculated the QW eigenenergies at the Γ point within the envelope-function formalism,[18,19] and deduced the 2D Rydberg energies of the heavy and light excitons from the theoretical calculation of Green *et al.*,[20] then we have calculated the transition energies. We assumed a valence-band offset $\Delta E_v = 0.31 \Delta E_g$ according to recent experimental findings[21,22] and the energy gap values $E_g(\text{GaAs}) = 1.519$ eV, $E_g(Ga_{0.75}Al_{0.25}As) = 1.815$ eV. The nonparabolic effective masses were calculated according to Kane's three-band model[23,24] with $m_e(\text{GaAs}) = 0.0665 m_0$ at the bottom of the well and $m_e(Ga_{1-x}Al_xAs) = (0.0665 + 0.0174x + 0.145x^2) m_0$ in the barrier. The hole effective masses have been expressed as a function of Luttinger parameters given by Lawaëtz,[25] linearly interpolated between GaAs and AlAs. Concerning the calculation of the hole eigenenergies, we have used for the band-extremum effective masses $m_{LH} = m_0/(\gamma_1 + 2\gamma_2)$, and $m_{HH} = m_0/(\gamma_1 - 2\gamma_2)$ in the well and $m_{LH} = m_{HH} = m_0/\gamma_1$ in the barrier. A self-consistent calculation gives for $L_z = 21$ atomic monolayers ($a/2 = 2.83$ Å), the electron and hole confinement energies $E_e = 58.4$ meV, $E_{HH} = 19$ meV, and $E_{LH} = 32.3$ meV. The 2D Rydberg energies deduced from Ref. 20 are, respectively, 8.70 and 8.74 meV for the heavy and light excitons. The corresponding transition energies are then $E(C_1H_1) = 1.588$ eV and $E(C_1L_1) = 1.601$ eV. The same calculation gives for $L_z = 20$ monolayers $E(C_1H_1') = 1.593$ eV and $E(C_1L_1') = 1.606$ eV, then $E(C_1H_1') - E(C_1H_1) = E(C_1L_1') - E(C_1L_1) = 5$ meV. This is in very good agreement with the experimental values and clearly shows that in our samples the pairs of structures result of interfacial defects of only one atomic monolayer.

Figure 3 shows the low-temperature differential reflectivity spectra of the structure obtained by wavelength-modulation (a) and piezomodulation (b) techniques. For clarity, the amplitude of the $E(C_1H_1)$ structure has been normalized to one in both spectra. The ratio $E(C_1L_1)/(C_1H_1)$ of the structures is K_W in WR and K_P in PR. Clearly, K_P and K_W are nonequal. In WMT, which is a nonselective modulation technique, the modulation parameter is the same for all transitions, so that the ratio $K = K_P/K_W$ gives directly the ratio of the piezomodulation parameters of the $E(C_1L_1)$ and $E(C_1H_1)$ transitions, we measure $K_{\text{exp}} = 2.6$. Figure 4 shows the same type of comparison for the 100-K

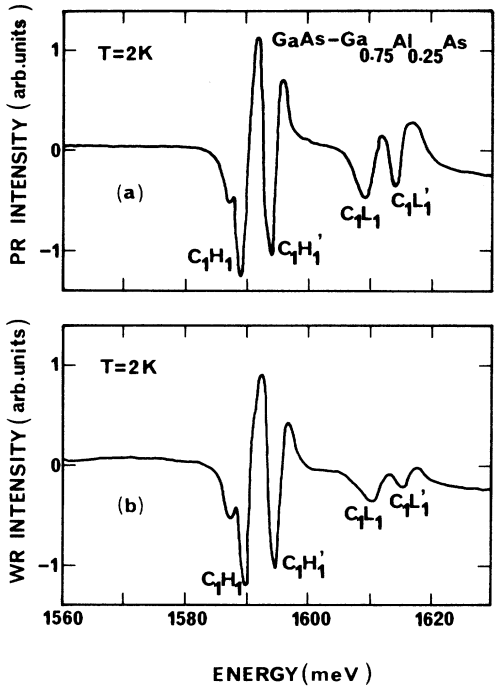

FIG. 3. Low-temperature differential reflectivity spectra obtained by wavelength-modulation (a) and piezomodulation (b) techniques. In both spectra the $E(C_1H_1)$ amplitude has been normalized to one.

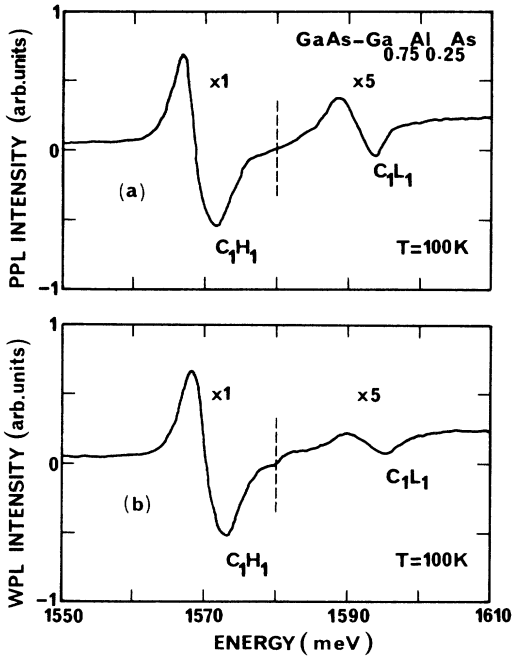

FIG. 4. 100-K differential photoluminescence spectra. (a) Wavelength-modulation spectrum. (b) Piezomodulation spectrum. In both spectra the $E(C_1H_1)$ structure has been normalized.

differential photoluminescence spectra, we measure the same value $K=2.6$. Here, one could expect additional $E(C_1H_1')$ and $E(C_1L_1')$ structures to appear. They actually do not, which we explain by noting the thermal enlargement of the PL lines slightly larger than the difference between the $E(C_1H_1)$ $E(C_1L_1)$ and $E(C_1H_1')$ $E(C_1L_1')$ states.

In piezomodulation technique, the exciton-state-energy modulation dE/dX is completely expressed in terms of deformation potentials of the Γ_6 conduction-band minimum and Γ_8 valence-band maximum.

The stress Hamiltonian[26,27,28] can be written as

$$H_e=-a(e_{xx}+e_{yy}+e_{zz})-3b[(L_x^2-\tfrac{1}{3}L^2)e_{xx}+\text{c.p.}]$$
$$-\frac{6d}{\sqrt{3}}[(L_xL_y)e_{xy}+\text{c.p.}]\ ,$$

where L is the angular-momentum operator, c.p. denotes cyclic permutation with respect to the indices x, y, and z, a is the hydrostatic deformation potential of GaAs band gap, and b and d are the shear deformation potentials of the Γ_8 valence band.

Under a (001) coplanar stress, the strain-tensor components e_{ij} are

$$e_{xx}=e_{yy}=(S_{11}+S_{12})x\ ,$$
$$e_{zz}=S_{12}x\ ,$$
$$e_{xy}=e_{yz}=e_{zx}=0\ ,$$

where S_{11}, S_{12} are the compliance coefficients.

The light- and heavy-hole states shift are, with respect to the Γ_6 conduction band with a rate, given by

$$\frac{dE_{\rm HH}}{dX}=2a(S_{11}+2S_{12})-b(S_{11}-S_{12})\ ,$$
$$\frac{dE_{\rm LH}}{dX}=2a(S_{11}+2S_{12})+b(S_{11}-S_{12})\ .$$

Using the values $S_{11}=1.16\times10^{-6}$ bar^{-1}, $S_{12}=-0.37\times10^{-6}$bar^{-1}, $a=-8.4$ eV, and $b=-1.76$ eV,[27] we obtain

$$\frac{dE_{\rm HH}}{dX}=-4.3\ \text{meV/kbar}\ ,$$
$$\frac{dE_{\rm LH}}{dX}=-9.7\ \text{meV/kbar}\ ,$$

so that the calculated ratio of amplitude modulation of the light-hole energy with respect to the heavy-hole one is given by

$$K_{\rm calc}=\frac{dE_{\rm LH}/dX}{dE_{\rm HH}/dX}=2.3\ .$$

We obtain a value very close to the experimental one deduced from the spectra given in Figs. 3 and 4. This permits us to unambiguously identify $E(C_1H_1)$ and $E(C_1L_1)$ as heavy- and light-hole transitions, respectively. On the other hand, the fact that $K_{\rm exp}\simeq K_{\rm calc}$ clearly shows that, in our structures, there is no light- and heavy-hole states mixing.

In conclusion, we have reported a differential spectroscopy investigation of good quality GaAs-GaAlAs quantum wells. We have shown for the first time that the piezomodulation technique is a powerful tool in order to identify heavy- and light-hole states in quantum wells and superlattices. Such an experimental investigation can be fruitfully extended to the case of other types of microstructures. It is worthwhile to note that the value of the K ratio, which is 2.3 in GaAs, is a function of the relative magnitudes of the hydrostatic and shear deformation potentials. In other semiconductors this value may be very different. As an example, in CdTe the shear deformation potential b is very important so that $dE_{\mathrm{HH}}/dx = -3.4$ meV/kbar and $dE_{\mathrm{LH}}/dx = +14$ meV/kbar. This gives for the K ratio the value -4.1. As a consequence, the light- and heavy-hole states are piezomodulated out of phase, and the identification is obvious.

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