Optical investigation of confinement and strain effects in CdTe/$Cd_{1-x}Zn_xTe$ single quantum wells

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Transmission, luminescence, and excitation spectra are presented for strained $Cd_{1-x}Zn_xTe$/CdTe/$Cd_{1-x}Zn_xTe$ quantum-well heterostructures grown by molecular-beam epitaxy with CdTe thicknesses ranging from 1000 to 50 Å. The effect of carrier confinement in such quasi-bidimensional systems is shown by substantial energy shifts of excitonic transitions related to the existence of quantized states. The structures are of mixed type with electrons and heavy holes confined in the CdTe wells (type I), whereas the light holes are localized in the $Cd_{1-x}Zn_xTe$ barriers (type II). These results also allow an estimation of the variation with thickness of the 2D-exciton binding energy.

I. INTRODUCTION

The study of strained-layer quantum-well heterostructures (SL QWH's) of lattice-mismatched materials has been widely developed for III-V semiconductors[1] whereas SL QWH's and strained-layer superlattices constructed with II-VI compounds have been grown and characterized only recently (for example, see Kolodziejski *et al.*[2] and references therein for superlattices made with II-VI–based manganese compounds, and Ref. 3 for CdTe-ZnTe superlattices). For II-VI SL QWH's, extensive optical studies have been carried out for the CdTe/$Cd_{1-x}Mn_xTe$ system (Refs. 4 and 5). With SL QWH's, it is possible to obtain high-quality heterostructures because the lattice mismatch is accommodated by elastic strain rather than by misfit dislocations at the interface, provided that the layer thicknesses are less than some critical value. Besides the possibility of enlarging the choice of epitaxial materials on a given substrate or buffer, the intrinsic interest of these structures is that one can tailor their optical and electronic properties by the combination of *two* physical effects, the band-gap discontinuity *and* the mismatch-induced strain.

In CdTe/$Cd_{1-x}Zn_xTe$ SL QWH's, three experimental parameters can change the electronic properties of the system. First, the choice of the buffer-layer composition determines the strain effect on the band configuration of the heterostructure. The energy shift of the conduction band ΔV_c, and the shifts and splitting of the valence-band extrema (ΔV_{hh} for the heavy holes and ΔV_{lh} for the light holes), can be written as

$$\Delta V_c = 2a_c(S_{11}+2S_{12})X ,$$
$$\Delta V_{hh} = 2a_v(S_{11}+2S_{12})X - b(S_{11}-S_{12})X , \qquad (1)$$
$$\Delta V_{lh} = 2a_v(S_{11}+2S_{12})X + b(S_{11}-S_{12})X ,$$

where a_c and a_v are the hydrostatic deformation potentials of the conduction and valence bands, respectively, b is the shear deformation potential of the valence band, S_{11} and S_{12} are the elastic compliance constants, and X is the in-plane stress experienced by the layers:

$$X = \frac{a_{subs}-a_l}{a_l}\frac{1}{(S_{11}+S_{12})} . \qquad (2)$$

a_{subs} (a_l) is the lattice parameter of the substrate (layer). The values of all the parameters involved in Eqs. (1) and (2) are deduced from the work of Thomas,[6] with the ratio a_c/a_v given by Camphausen *et al.*[7] In our case, CdTe/$Cd_{0.92}Zn_{0.08}Te$ SL QWH's grown on $Cd_{0.96}Zn_{0.04}Te$, the CdTe layer is in biaxial compression ($X<0$) and the heavy holes are pushed above the light holes, whereas the $Cd_{1-x}Zn_xTe$ barriers are in biaxial dilatation ($X>0$) so that the valence-band maximum in the barriers is defined by the light holes (Fig. 1). Second, by varying the thickness of the quantum-well layer it is possible to control the

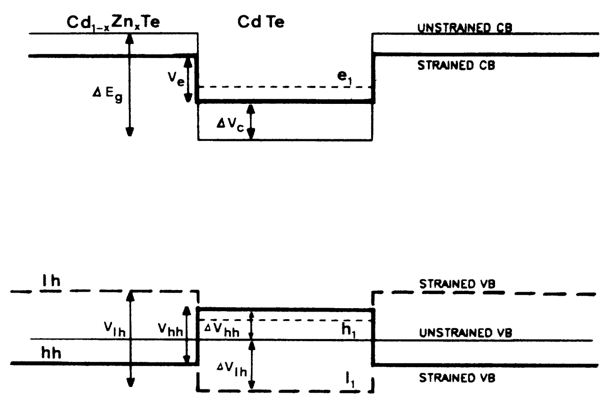

FIG. 1. Schematic diagram of the conduction and valence bands of a strained CdTe/$Cd_{1-x}Zn_xTe$ double heterostructure, before (thin solid line) and after (heavy dashed and heavy solid lines) including the effect of strains, assuming no valence-band offset in the absence of strains. The thin dashed lines in the well represent the ground states for electrons (e_1) and heavy holes (h_1).

importance of the confinement effect for electrons and holes in the well. Third, by changing the alloy composition x in the barriers, the band-gap discontinuity ΔE_g can be adjusted. The partition coefficient α of ΔE_g between the conduction and valence bands has been measured for the CdTe/ZnTe system giving $\alpha = 0.87$ (Ref. 8) and more recently $1.05 \leq \alpha \leq 1.20$ (Ref. 9). With a valence-band offset of zero ($\alpha = 1$), the electronic band structure of the SL-QWH's studied here would be as shown in Fig. 1. The strain field in the SL-QWH completely determines the potential wells for the holes whereas it only reduces the well for the electrons confined in the CdTe layers. Depending on the three parameters (buffer-layer, well thickness, alloy composition) the system can be of type I for the electron–heavy-hole exciton recombination and type II for the electron–light-hole one as seen in Fig. 1.

In this paper, we report on CdTe/$Cd_{1-x}Zn_xTe$ SL QWH's with the band configuration of Fig. 1. Experiments are described in Sec. II. Photoluminescence, transmission, and excitation data are presented in Sec. III for a set of samples with confined CdTe layers of different thicknesses. In Sec. IV, all the spectroscopic observations are related to the combined effects of band-gap offsets and lattice mismatch on the band structure of strained layers. From this discussion, exciton binding energies are estimated and found to vary appreciably with the CdTe thickness.

II. EXPERIMENTS

Our samples were grown by molecular-beam epitaxy (MBE) on (100)-oriented $Cd_{1-y}Zn_yTe$ substrates with $y = 2.5-4\%$. They consist of one CdTe layer confined between two 800-Å $Cd_{1-x}Zn_xTe$ barriers with x of the order of 8%. The layers are not intentionally doped. Nominal thicknesses L_w of the CdTe well layer are 1000, 450, 200, 100, and 50 Å. The thicknesses were obtained from secondary-ion mass spectroscopy profiling studies and by high-resolution transmission electron microscopy. These are known with an accuracy of 10%. The thickness of the whole structure is in all cases less than the critical value at which the strain relaxes.[10] Therefore in this set of samples, the $Cd_{0.92}Zn_{0.08}Te$ barriers and the CdTe layer are uniformly strained in an opposite manner, due to the $\simeq 0.23\%$ lattice-parameter mismatch with respect to the $Cd_{0.96}Zn_{0.04}Te$ substrate. Luminescence was excited with a krypton laser or with a dye laser (LD700 dye). A tungsten lamp was used for the transmission measurements. All the experiments were performed at 1.8 K.

III. SPECTROSCOPIC RESULTS

Figure 2(a) presents the transmission results obtained for a set of samples with varying CdTe quantum-well thicknesses. This technique, which involves resonant absorption of exciting light, is possible when $L_w \geq 100$ Å, in which case the onset of the substrate band-edge absorption lies at higher energy than the quantum-well states. The main resonance corresponds to the ground state of the free exciton in the well, i.e., the electron–heavy-hole e_1h_1 absorption. The shift of the e_1h_1 transition to higher energies for narrower quantum wells illustrates clearly the importance of the confinement effect for the electrons and the heavy-holes in the CdTe layers. However, the effect of the confinement is partially reduced by the increase of the 2D heavy-exciton binding energy when the L_w thickness is decreased. For the 1000-Å-thick layer, the structures which are observed at higher energy than the exciton ground state correspond to the oscillations attributed to quantized states of the exciton polariton.[11] The feature at 1609 mV involves the electron–light-hole exciton in the confined layer with an energy splitting between light- and heavy-hole excitons in

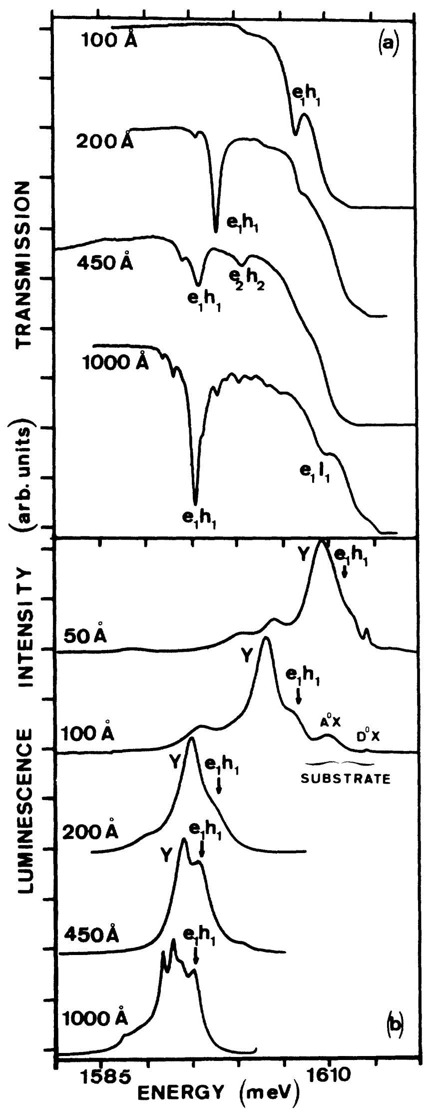

FIG. 2. (a) Band-edge transmission spectra (1.8 K) of $Cd_{1-x}Zn_xTe$ quantum-well heterostructures showing a clear energy shift of the e_1h_1 heavy-exciton resonance in the well, as a function of the CdTe layer thickness. (b) Photoluminescence spectra (1.8 K) of the same set of quantum-well samples. The arrows correspond to the transition of the exciton ground state in the wells as deduced from the transmission spectra [(a)].

agreement with the splitting observed for an unrelaxed CdTe layer on a $Cd_{0.96}Zn_{0.04}Te$ substrate.[10] The 450-Å-well's transmission spectra exhibit, at higher energy than the e_1h_1 main excitonic transition, a structure at 1601 meV which we attribute (see later) to the quantized state e_2h_2 of the exciton in the CdTe quantum well.

Considering the photoluminescence (PL) spectra [Fig. 2(b)], we first mention that the luminescence peaks are more intense than in the case of a 2-μm-thick CdTe MBE layer. For the 1000-Å-thick layer, as well as the very strong e_1h_1 intrinsic emission as already observed for similar samples,[12] we observe lower energy lines which are attributed to excitons bound to residual donors and acceptors. For the thinner layers (450–50 Å), the emission spectra due to the SL-QWH are dominated by a peak labeled Y which is shifted down in energy by $\simeq 3$ meV from the peak of the e_1h_1 excitonic ground-state line seen in the transmission spectra. The PL maximum Y corresponds to the small feature seen on the low-energy side of the intrinsic exciton resonance in the transmission spectra [Fig. 2(a)] whereas the latter appears in the PL spectra as a shoulder of the Y line.

We discuss now the possible interpretations for the Y line. From their energy positions compared to those of the intrinsic excitons, these emission lines Y appear at first to correspond to extrinsic excitons bound to donors in the confined CdTe layers (the binding energy of excitons bound to residual donors in bulk CdTe is 3 meV). However, we have to consider the following facts. (i) The Y line intensity compared to the e_1h_1 intensity increases when the thickness is reduced (note also that the corresponding feature in the transmission spectra does not decrease with L_w). (ii) The expected saturation effect for extrinsic emission was not observed in the PL spectra when the laser intensity was increased by a factor of 100. Moreover, it was not possible to observe two-electron transitions in the emission spectra by selectively exciting the Y line, in contrast with the 2-μm-thick MBE CdTe layers.

All these observations suggest that the Y lines correspond mainly to intrinsic exciton recombinations. In $GaAs/Ga_{1-x}Al_xAs$ quantum wells, such energy shifts between the main absorption and emission exciton lines have been observed[13] and attributed to processes involving intrinsic excitons migrating to lower energy states localized on random interface defects. This is a reasonable explanation for the Y line but we should also mention another interpretation. The energy position of line Y corresponds to that expected in our system for the type-II transition between the electrons confined in the CdTe conduction-band wells and the light-hole ground state in the barriers (Fig. 1). In both interpretations, the Y-line transition is expected to have a low oscillator strength: this is due to the low density of states in the former case and to the indirect nature of the recombination in the latter. However, in emission, both processes could be efficient as a result of exciton mobility and energy-transfer effects. What could distinguish the two interpretations is to check experimentally if light holes are involved in the Y transition. Preliminary results of piezomodulated reflectivity performed on the same samples indicate a light-hole transition for the Y line,[14] favoring the indirect, type-II transition model. However, optical-pumping experiments performed by modulating circularly polarized light show that the Y transition seen in absorption is mainly due to a heavy hole.[15] Thus it seems that the Y line corresponds to two (or more) recombination channels which are difficult to separate. The low value of the barrier-height for the valence band which probably leads to large admixture of the hole states in the well and in the barriers could be the origin of these difficulties. Growth of structures with different band configurations are in progress and should give additional information concerning the exact nature of the Y recombination mechanism in $CdTe/Cd_{1-x}Zn_xTe$ heterostructures.

Figure 3 presents a typical excitation spectrum for the 450-Å confined layer, detected at the maximum of the Y line. This spectrum exhibits two clearly resolved peaks at 1596.4 and 1601 meV involving the excitonic transitions in the well, and two others at higher energies (1625 and 1639 meV) which correspond to free-exciton transitions in the barriers. The position of these two high-energy lines, their energy splitting, and their relative intensities (the high-energy line is three times stronger than the low-energy one) clearly indicate they are due to energy transfer from light-hole excitons (1625 meV) and heavy-hole excitons (1639 meV) created in the barriers. The reversal of the light- and heavy-hole-exciton transitions is a striking consequence of the tensile stress experienced by the $Cd_{1-x}Zn_xTe$ barriers. The exciton transitions in the well correspond exactly to the structure observed in transmission spectra [Fig. 2(a)], and are attributed to the exciton ground state e_1h_1 and to the quan-

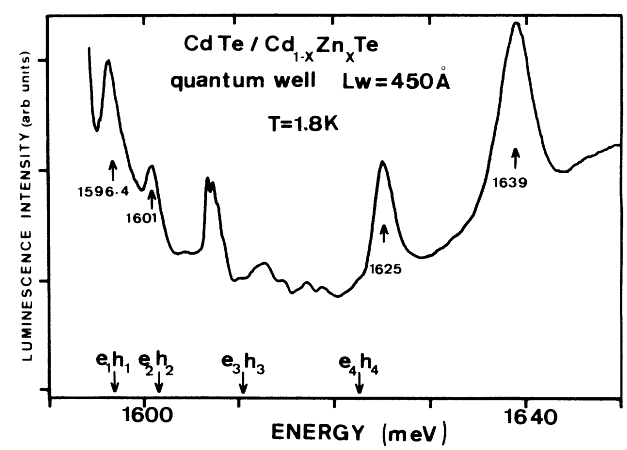

FIG. 3. Typical excitation spectrum of $CdTe/Cd_{1-x}Zn_xTe$ quantum-well heterostructures for a 450-Å-thick CdTe layer with detection at 1593.6 meV corresponding to the Y emission maximum. The two transitions at 1596.4 and 1601 meV are attributed to the exciton ground state e_1h_e and to the exciton quantized state e_2h_2 in the well whereas the two peaks on the high-energy side are due to light-hole (1625-meV) and heavy-hole (1639-meV) exciton transitions in the barriers. The solid arrows along the energy axis indicate the calculated transition energies involving heavy-hole states as discussed in the text.

tized state e_2h_2 in the CdTe confined layer. The structure at 1.606 eV is deduced to be mainly due to resonance with the donor-bound exciton of the substrate by comparison with PL and excitation spectra of the sample rear face and the dip at 1609 meV corresponds to the substrate intrinsic exciton. First results of piezomodulated reflectivity spectra and of optical-pumping data with polarized light confirm the identification of the different contributions of light-hole and heavy-hole states.[14]

IV. DISCUSSION

As the total thickness of the SL QW is smaller than the critical value, all the layers are unrelaxed with the strained state defined by the lattice-parameter mismatch. From the energy positions (as well as the splitting) of the barrier's intrinsic exciton transition, we obtain an estimate of the actual barrier alloy composition x, using Eqs. (1) and (2). For the 450-Å SL QWH shown in Fig. 3, $x=0.068$ with a substrate alloy composition y checked to be 0.025. Then we deduce the band-gap discontinuity ΔE_g which, with $\alpha=1$, is the conduction-band offset, and from Eq. (1), the strain contribution for all the bands in both the CdTe well and the $Cd_{1-x}Zn_xTe$ barriers. For example, in the 450-Å SL QWH, the different potential wells V_e, V_{hh}, and V_{lh} (see Fig. 1) are $V_e=32.4$ meV, $V_{hh}=7.34$ meV, and $V_{lh}=-15.2$ meV (in all the samples studied here the value of V_{lh} is negative, corresponding to the light-hole ground state's lying in the $Cd_{1-x}Zn_xTe$ barriers). With a finite-barrier quantum-well model, using for the effective masses of electrons and holes in the CdTe the values $m_e=0.088m_0$ and $m_h=0.6m_0$ (Ref. 16), the confinement energies E_{e_n} (E_{h_n}) for the nth electron (heavy-hole) quantum state could be estimated. Then the energy for an exciton transition related to the nth electron quantized state and the nth heavy-hole state is

$$E_{e_nh_n}=E_{g_{\mathrm{CdTe}}}-\Delta V_{\mathrm{hh}}+\Delta V_c+E_{e_n}+E_{h_n}-E_{\mathrm{bh}} , \quad (3)$$

where E_{bh} is the binding energy of the heavy-hole exciton in the CdTe confined layer. Comparison of the experimental values of the e_1h_1 excitonic transitions with Eq. (3) leads to an estimate of E_{bh} as a function of L_w. The results are reported in Table I. The exciton binding energy increases markedly when the well thickness L_w decreases, as expected in theoretical confinement models[17] and as observed in III-V heterostructures.[18] The whole calculation for the potential wells and the carrier energy levels has been carried out with different values of the coefficient α, that is, with different ratios of offsets for conduction and valence bands. The results are reported in Fig. 4, which compares the calculated optical gap

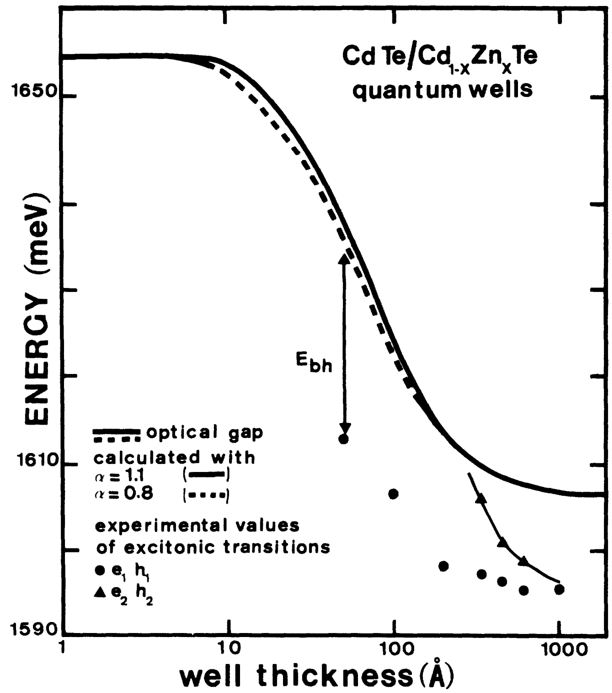

FIG. 4. Comparison of the calculated optical gap of a $CdTe/Cd_{1-x}Zn_xTe$ single quantum well vs the experimental value of the excitonic transition e_1h_1 as a function of the CdTe well thickness. The optical gap is calculated with a one-dimensional rectangular well model having a finite depth and for varying values of α. The difference E_{bh} between the optical and excitonic gaps allows us to deduce the values of the exciton binding energies E_{bh}. The experimental values of e_2h_2 exciton transitions are reported (▲) and compared to the calculated curve (thin solid line).

TABLE I. Experimental values of exciton transition energies (e_1h_1) and zinc composition in the structures for different CdTe well thicknesses (L_w = 1000, 450, 200, 100, and 50 Å). V_e and V_{hh} are the potential wells used to calculate the confinement energy of electrons (E_{e_1}) and holes (E_{h_1}) with $\alpha=1$. The exciton binding energies E_{bh} are obtained from Eq. (3) as explained in the text.

	1000 Å	450 Å	200 Å	100 Å	50 Å
e_1h_1(expt) (meV)	1595.4	1596.4	1597.8	1606.6	1612
x% Zn barriers	9.4	6.8	8.4	8.5	8.2
y% Zn substrate	4.3	2.5	4.2	3.8	3.6
V_e (meV)	45.2	32.3	40.2	40.6	39.2
V_{hh} (meV)	10.1	7.3	9.1	9.2	8.8
E_{e_1} (meV)	0.38	1.56	5.97	14.76	26.15
E_{h_1} (meV)	0.06	0.24	0.97	2.59	5.10
E_{bh} (meV)	11	12	15.3	17	25.4

(e_1h_1) as a function of the well thickness with the experimental data. The difference between the experimental e_1h_1 exciton transition and the calculated optical gap is attributed to the exciton binding energy E_{bh}. For a 50-Å-thick CdTe well layer, the value of E_{bh} is found to be more than two times larger (25 meV) than that of the three-dimensional (3D) exciton. A similar value has been obtained recently for a 50-Å-thick CdTe/(Cd,Mn)Te multiquantum well.[5] We emphasize that, when we vary α from 0.8 to 1.1, the value of E_{bh} is changed by only 4% for the 50-Å CdTe well (Fig. 4). That is, determination of the 2D exciton binding energy is possible in these systems because the variations of the confinement energies E_{e_1} and E_{h_1} when α is changed are relatively small compared to the value of E_{bh}. The accuracy in the value deduced for E_{bh} is then mainly dependent on the precise determination of the deformation potentials a_c, a_v, b in CdTe and ZnTe [see Eq. (1)]. We would like to mention also that a value of $\alpha=1.2$ corresponds to a band configuration where both the heavy- and light-hole ground states are localized in the barriers, which would give rise to heterostructure with indirect transitions (type II). This is in contradiction with the experimental results which exhibit a high absorption oscillator strength for the e_1h_1 transition in these structures. As a consequence, values $\alpha \geq 1.2$ for the partition coefficient in the system $CdTe/Cd_{1-x}Zn_xTe$ can be ruled out.

Once the exciton binding energies are known, Eq. (3) can be used to calculate transition energies for other quantum states. For a given well thickness, the results are indicated in Fig. 3 by the solid arrows along the energy axis. The small shift between the calculated position of e_2h_2 and the line attributed to this transition can be due to the 10% uncertainty in the thickness. As far as the variation with well thickness L_w is concerned, Fig. 4 presents experimental results for the e_2h_2 energy position (▲) compared to the calculated curve (fine solid line). The good agreement between experimental and calculated values of the variation of the e_2h_2 energy as a function of L_w confirms the validity of our approach to the analysis of the spectroscopic data obtained on these strained quantum-well structures. The nonobservation of the e_2h_2 transition for thinner wells ($e \leq 200$ Å) is due to the superposition of either substrate or barrier transitions in the energy range where e_2h_2 is expected.

In conclusion, we have studied the optical properties of $CdTe/Cd_{1-x}Zn_xTe$ quantum wells. In order to keep good crystal quality and to work on an unrelaxed system where the actual strain state is known, the Zn concentration of the barriers has been kept below 20% and single quantum wells have been grown. In this case, sharp lines in emission and absorption as well as good radiative efficiency have been obtained. While the well depth is relatively low (40 meV for the electrons), the confinement and the quantization of the carrier motion have been detected. The strong e_n-h_n ($n \leq 2$) transitions observed in absorption and excitation spectroscopy show that these structures are of type I as a result of both the band offset and the strain on the electronic-band-gap configuration. These results indicate that the partition coefficient α between CdTe and ZnTe is less than 1.2 in agreement with other determinations. The difference between the experimental position of the e_1-h_1 exciton transitions and the calculated optical band gap has been attributed to the variation of the exciton binding energy with the layer thickness. Recombination processes are more complicated. Whereas the fundamental e_1-h_1 transition is observed in emission, the luminescence spectra are dominated at low excitation density by a line Y at lower energy. Possible explanations are exciton recombinations on interface defects or transitions between electrons in the well and holes in the barriers. So far the experimental data cannot provide a definite interpretation. The situation is made difficult by the low value of the valence-band shift at the interface. It is clear that the direct observation of the type-II transition in excitation spectra is essential to have a precise measurement of the valence-band offset between CdTe and ZnTe.

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