Photoluminescence studies of planar-doped GaAs-$Ga_{1-x}Al_xAs$ multiple-quantum-well structures

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Photoluminescence experiments on various selectively silicon "δ-doped" GaAs-$Ga_{1-x}Al_xAs$ multiple quantum wells, grown by molecular-beam epitaxy, have been performed. For comparison, photoluminescence of a similar undoped structure and of silicon "δ-doped" bulk GaAs were studied. The assignment of the luminescence structures was obtained through a careful analysis of the temperature, excitation power, and magnetic-field dependence of the spectra. As expected, although most of the silicon atoms behave as donors, acceptor states related to this impurity have been detected. Due to the small doping level of the samples, it turns out that the amount of Si acceptor states is comparable to that of the residual carbon acceptor states, allowing a direct comparison between the related densities. It is found that some of the silicon dopants are located where they were intended to be placed during the doping procedure. However, a significant number of Si impurities are found to be shifted in the direction of growth with respect to their intended locations, in agreement with magnetotransmission measurements performed on the same samples. These investigations provide interesting information about the self-compensation and segregation of impurities during the molecular-beam epitaxy of the samples.

I. INTRODUCTION

Shallow impurities play a dominant role in determining the optical and transport properties of semiconductors. In the case of quasi-two-dimensional quantum-well structures, which can be obtained by modern growth techniques such as molecular-beam epitaxy (MBE), these properties depend strongly on quantum-well dimensions and the position where the impurities are located, since they change their binding energies with this location.[1-4]

In the past few years there has been a growing interest in selective doping, characterization, and fundamental properties of such structures.[5-11] There are several important properties that can be studied in such systems by optical methods: (i) the influence of the impurities on the radiative recombination processes in quasi-two-dimensional (2D) systems, (ii) the differences between two- and three-dimensional (3D) systems, and (iii) the characterization of intentionally doped structures.

To address these problems we have performed photoluminescence experiments on various, selectively silicon doped GaAs-$Ga_{1-x}Al_xAs$ multiple quantum wells (MQW) grown by MBE and previously studied in magnetotransmission experiments.[10] These results are compared to those obtained on a similar "δ-doped" bulk GaAs sample. The samples and experimental procedure are described in Sec. II. In Sec. III the experimental results are presented. The wide temperature and excitation power ranges investigated, as well as experiments in magnetic fields, allow us to interpret the structures we observed. A detailed discussion of the variation of the recombination processes as a function of the doping profile, the excitation power, and the temperature are presented in Sec. IV. The conclusions appear in Sec. V.

II. SAMPLES AND EXPERIMENTAL PROCEDURE

Selectively Si doped GaAs-$Ga_{1-x}Al_xAs$ ($x=0.25$) MQW (150 periods) were grown by MBE. The wells and barriers were chosen to be 100 Å thick. The intended doping profile was of the "δ-type," with growth interruption during the doping process. This realizes the so-called "stop and go" method.[12] The procedure first involves the growth of a 7000-Å-thick GaAs buffer, then that of a 5000-Å-thick barrier of $Ga_{0.75}Al_{0.25}As$ followed by the MQW sequence, which in turn is covered by a 50-Å GaAs cap layer. The temperature of the substrate has been kept constant during the whole process at a relatively high level ($T_s=640$ °C). This temperature ensures very good growth conditions for undoped layers, as this can be confirmed by the luminescence properties of the MQW or the very high mobilities of the heterojunctions obtained this way.[13] However this temperature may not be optimized to avoid segregation processes when doping the samples.[14] Three samples were prepared this way, with a doping spike (with $N_d=10^{10}$ cm^{-2}) located at the beginning of the well, 10 Å away from the interface (sample 1), in the middle of the well (sample 2), and at the end of the well, 10 Å before the next interface (sample 3). For comparison, similar MQW, uniformly doped ($N_d=9.9\times10^{15}$ cm^{-3}) (sample 4) and undoped (sample 5), as well as δ-doped bulk GaAs (sample 6) were investigated. The low

doping level, confirmed by transport measurements at 300 and 77 K, allows us to consider the impurities as isolated.[15] Typical Hall mobilities of the doped samples are 5000 and 25 000 $cm^2\ V^{-1}s^{-1}$ at 300 and 77 K, respectively.

Photoluminescence experiments have been performed in the temperature range 1.5–300 K using fiber optics and a standard cryostat with an He-gas-exchange cooling system. The very good reproducibility of the optical configuration gave us an opportunity to compare the results obtained for different samples and different experimental conditions. The photocarriers were excited using the 514.5-nm line of an Ar^+ laser or the 647.1-nm Kr^+ laser radiation. An excitation power density in the range 10^{-5}–10^2 W/cm^2 was used. The luminescent light was analyzed with a triple additive Raman spectrometer Dilor RTI30, followed by a cooled photomultiplier, operating in a photon counting mode. Magnetoluminescence experiments have been performed in a 13-T Bitter magnet.

III. EXPERIMENTAL RESULTS

A. Undoped multiple quantum well

In Fig. 1 the luminescence spectra obtained for undoped MQW (sample 5) measured at 4.2 K for several power densities are shown. For the lowest power, a single 1S heavy-hole free exciton line X is observed with a half-width at half maximum of $\Gamma=0.6$ meV. The energy peak position (1.5443 eV) is in agreement with the already published results for 100-Å GaAs-$Ga_{1-x}Al_xAs$ wells.[16,17]

Upon increasing the excitation power density, a second line can be observed at lower energy. This line disappears with increasing temperature. Such behavior allows us to assign this second line to a biexciton with a binding energy $E_B=1.0$ meV with respect to the free exciton, in good agreement with theoretical calculations[18] and previous experimental results.[19] The narrow, single 1S heavy-hole exciton line, observed in the undoped sample for the lowest excitation power shows the high quality of the samples: The samples have an equal width for the different wells and a small concentration of residual dopants.

At higher temperatures, the excited states, a 2S heavy-hole exciton and a 1S light-hole exciton, can be observed. The magnetic-field dependence of the excitonic transition energy, measured at 27 K, is shown in Fig. 2. Our results are in qualitative agreement with recent theoretical calculations which take into account the complex structure of the valence band.[20,21] These theoretical results are, however, not directly applicable to our case, since they are performed for a single quantum well, whereas our experimental results are obtained with a MQW. Nevertheless, the two-dimensional approximation[22,23] can be considered sufficient to describe the diamagnetic shift of 1S and 2S heavy-hole excitons as illustrated in Fig. 2 by solid lines. These theoretical results have been obtained by fitting the effective Rydberg and effective excitonic mass. In this simple model, the binding energy E_X of the 1S heavy-hole exciton is found to be equal to 7.9 ± 0.1 meV. This value is, as expected, smaller than the value theoretically predicted for a single quantum well:[21,24]

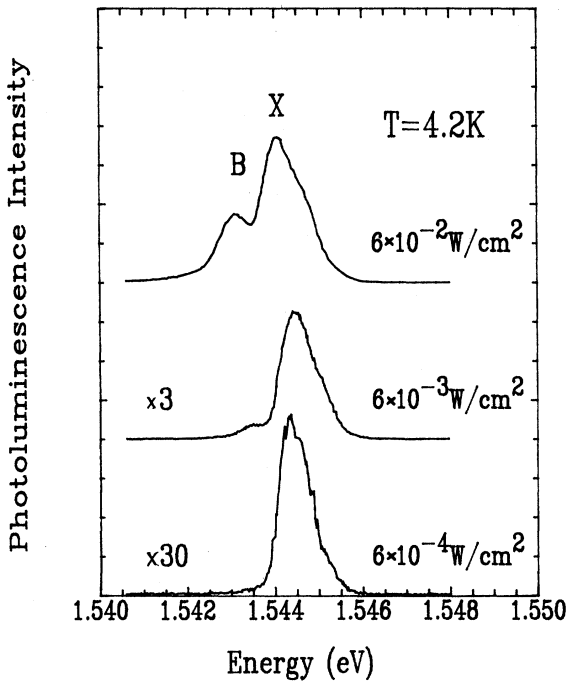

FIG. 1. Photoluminescence intensity as a function of the photon energy for sample 5 (undoped MQW), measured at 4.2 K with the indicated excitation power of the 514.5-nm radiation. The $n=1$, 1S heavy-hole exciton and biexciton are marked by X and B, respectively.

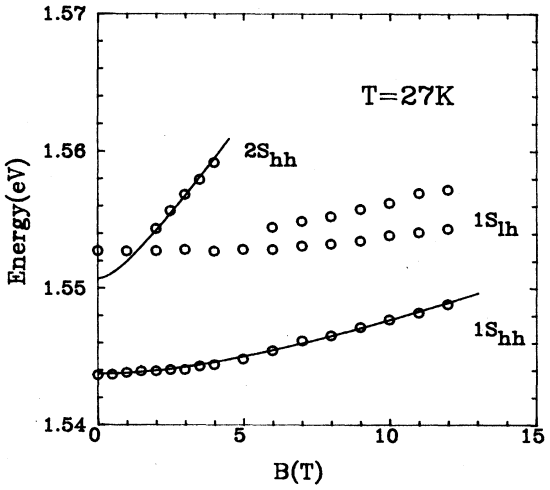

FIG. 2. Energy of the heavy-hole ($1S_{hh}$ and $2S_{hh}$) and light hole ($1S_{lh}$) excitons for sample 5 as a function of the magnetic-field strength, obtained from the photoluminescence experiment at 27 K (circles). The solid line is the calculated results for heavy-hole excitons in a two-dimensional, single-valence-band approximation.

This can be compared to the analogous results obtained for a donor state in a single and multiple quantum well (Fig. 1 in Ref. 3).

B. δ-doped and uniformly doped MQW

In Fig. 3 the power dependences of the luminescence spectra, measured at 4.2 K for sample 2 (MQW doped in the center), are presented. The free exciton line X is observed at 1.5446 eV. We assign the second dominant line (1.5429 eV) to an exciton bound to a neutral donor transition (D^0-X). This will be discussed in Sec. IV. The comparison of the photoluminescence spectra obtained for samples with different doping location is made in Fig. 4 for a power density of 3×10^{-3} W/cm^2. The luminescence intensity has been normalized to the peak intensity of the free exciton line X. The spectrum obtained for sample 1 (doped at the beginning of the well) is shifted by about 1 meV in comparison with the others, probably due to a slightly different quantum-well thickness for this sample. One can see that the lineshape of the spectrum obtained for sample 3 (doped at the end of the well) differs significantly from those obtained for the others. This spectrum is, in particular, broader and not well resolved.

In Fig. 5 the evolution of the luminescence spectrum with increasing temperature is shown for the uniformly doped MQW (sample 4). At 1.5 K the free exciton line X (1.5452 eV) and the exciton bound to a neutral donor (D^0-X) (1.5434 eV) are observed. The small structure around 1.515 eV is connected with excitonic transitions in the GaAs substrate. At a higher temperature (20 K), the intensity of the (D^0-X) line decreases, which is related to the ionization of the neutral donors. At the same temperature, two additional features, related to free-electron–neutral-acceptor transitions (e-A^0), are observed, and can be related to silicon and carbon acceptors. The presence at this temperature of 2D free electrons is revealed by the cyclotron resonance in far-infrared experiments performed on this sample (Fig. 3 of Ref. 10). The energy (1.523 eV) of the (e-C^0) transition is in agreement with those reported for a carbon acceptor in the MQW.[25] The second line (e-Si^0) reveals the amphoteric character of the Si impurities in GaAs and, to our knowledge, has not been observed earlier in MQW. Whereas the relative strengths of the two transitions involving both acceptor states are temperature dependent, they do not vary significantly with the power density at least for low values of the density.

At still higher temperatures (Fig. 5, 40 K), impurity-related transitions begin to disappear, due to the ionization of both acceptors and donors. The magnetic-field dependence of the acceptor-related features of the luminescence is presented for sample 4 (uniformly doped) in Fig. 6. In the quantum limit, $B > 2$ T ($\hbar\omega_c > kT$), the energies of the (e-C^0) and (e-Si^0) transitions increase

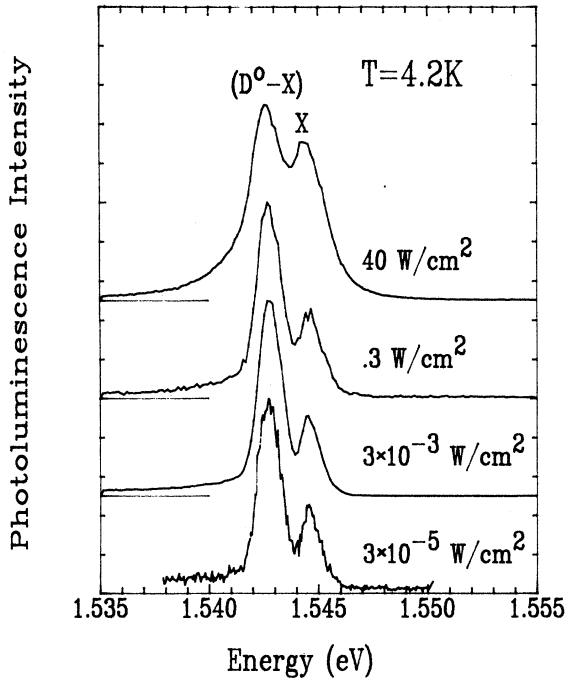

FIG. 3. Photoluminesence intensity as a function of the photon energy for sample 2 (doped in the well center) measured at 4.2 K, with the indicated excitation power of the 647.1-nm radiation. An effect of saturation of the exciton bound to the neutral donor line (D^0-X), in comparison with the 1S heavy-hole free exciton X, is observed for the highest excitation power.

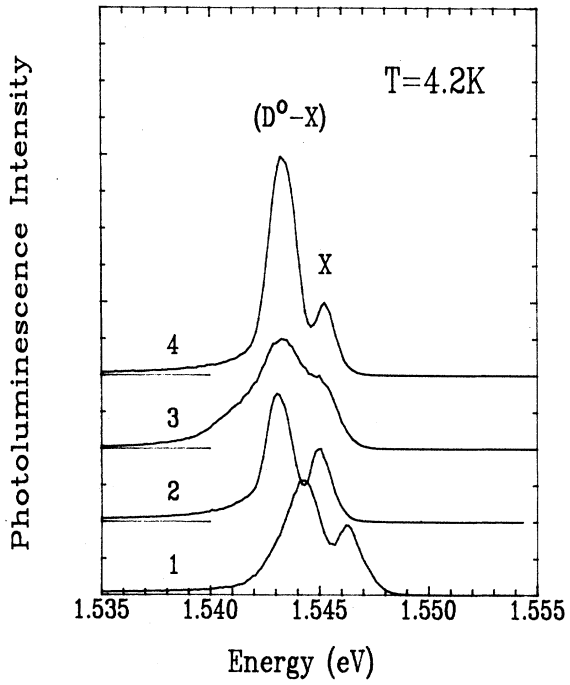

FIG. 4. A comparison of the photoluminescence spectra of samples 1–4 measured at 4.2 K. The excitation power is 3×10^{-3} W cm^{-2}. The spectra have been scaled and adjusted to the peak intensity of the free exciton line X in such a way that a direct comparison of the exciton bound to the neutral donor line (D^0-X) can be made.

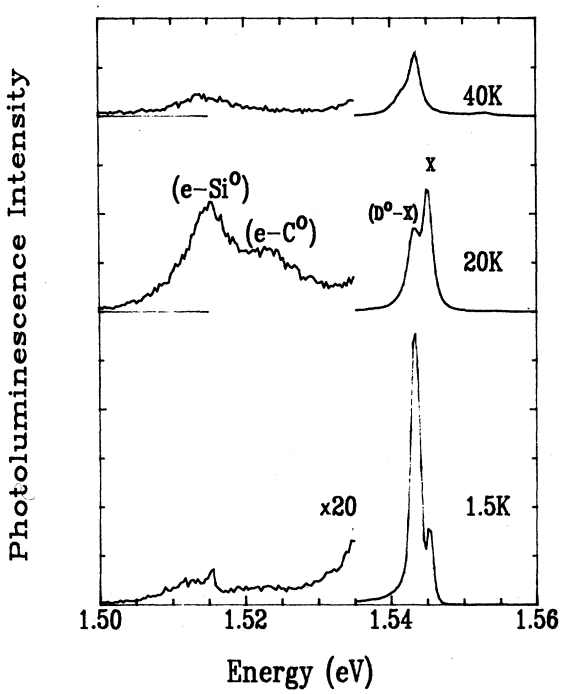

FIG. 5. Photoluminescence intensity as a function of the photon energy for sample 4 (uniformly doped), measured at different temperatures, excited by the 514.5-nm radiation. The power density is 3×10^{-3} W cm^{-2}. The four peaks at 20 K are identified as follows: $n=1$, $1S$ heavy-hole free exciton X; $n=1$, $1S$ heavy-hole exciton bound to the neutral donor (D^0-X); free-electron–neutral-acceptor recombinations (e-Si^0) and (e-C^0) for silicon and carbon acceptors, respectively.

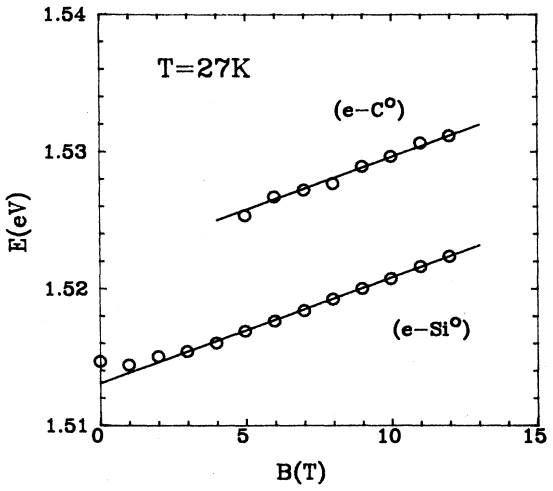

FIG. 6. Energy of the free-electron–neutral-acceptor transitions for the carbon (e-C^0) and silicon (e-Si^0) acceptors (circles) as a function of the magnetic-field strength, obtained from the photoluminescence experiment at 27 K (sample 4 uniformly doped). The solid lines display a linear dependence with a slope of $\frac{1}{2}\hbar\omega_c$.

linearly with the magnetic-field strength, with the same slope of 0.79 meV/T, which is in very good agreement with the value of $\frac{1}{2}\hbar\omega_c=0.78$ meV/T, obtained from the cyclotron resonance line position (Fig. 3 in Ref. 10). This corresponds to an effective mass m_e^* of the two-dimensional free electrons of $0.073m_0$.

In the range of magnetic fields up to 12 T, both X and (D^0-X) transitions show a typical excitonic character, with a constant energy difference of 1.8 ± 0.1 meV. Their diamagnetic shift can be described using the same parameters as those used for reproducing the results on sample 5 (Fig. 2).

C. δ-doped bulk GaAs

In Fig. 7 the temperature variation of the luminescence spectra obtained for sample 6 (Si δ-doped GaAs grown by MBE) is presented. In contrast to the MQW case, the spectrum is dominated at low temperature (1.5 K) by Si_{Ga}-Si_{As} (1.483 eV) and Si_{Ga}-C_{As} (1.491 eV) donor-acceptor pairs recombination. In the excitonic range, one can recognize[26] an exciton bound to a neutral donor (D^0-X) (1.5141 eV), an exciton bound to the neutral acceptor (A^0-X) (1.5124 eV), and a weak shoulder corresponding to the free exciton line X (1.515 eV).

With increasing temperature (20 K), a small decrease in the energies of the excitonic lines is observed that reflects a change of the GaAs energy gap. At the same time, the well-known evolution[27] from donor-acceptor

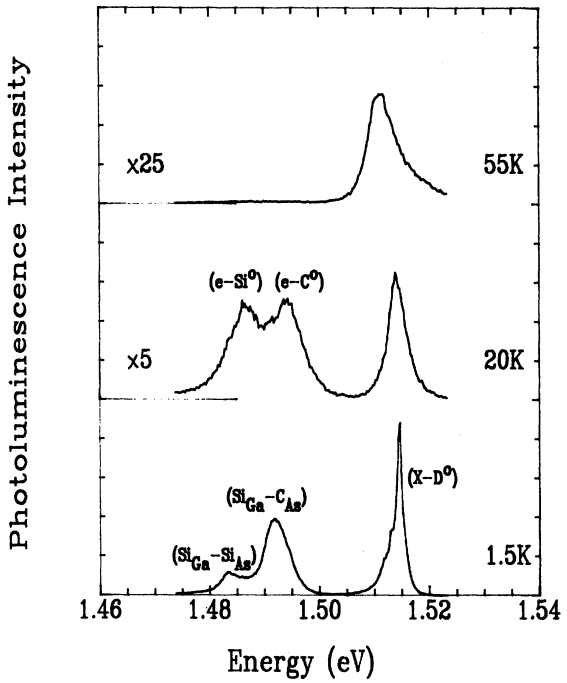

FIG. 7. Photoluminescence intensity as a function of the photon energy for sample 6 ("δ-doped" GaAs"), measured at different temperatures, excited by the 514.5-nm radiation. The power density is 3×10^{-3} W cm^{-2}. The identification and the evolution of the structures observed are described in the text.

pair recombination to free-electron–neutral-acceptor recombination is observed, manifesting itself in an increase by about 3 meV of the related-luminescence energies. At 55 K, due to the impurity ionization, only band-to-band luminescence, with a relatively small intensity, is found. The same evolution as that described above, from the donor-acceptor to free-electron–neutral-acceptor transition, is observed upon increasing the excitation laser power.

IV. DISCUSSION

In Sec. III systematic studies of the different MQW samples have been presented. Experiments performed as a function of temperature, excitation power, and external magnetic field showed that, in the n-type doping range investigated ($\approx 5\times 10^{15}$ cm^{-3}), at low temperatures ($T<30$ K), the luminescence in MQW is dominated by free exciton X and donor related lines. In our experiments, the luminescence intensity of both lines (Fig. 3) increases linearly with the excitation power, and the observed line shape of the spectra does not depend on the excitation power in the range of 3.10^{-5}–0.3 W/cm^2. However, as already reported in Ref. 5, for a higher excitation power (0.3–40 W/cm^2) the line shape changes significantly due, in our opinion, to the saturation of the donor related line. From such behavior, it is concluded that the same number of photoexcited particles are involved in both recombination processes. Thus the lower energy line is assigned to an exciton bound to a neutral donor (D^0-X). This interpretation is confirmed by the results of experiments performed as a function of the magnetic field and the temperature and it agrees with that given recently by Liu *et al.*[9] This contradicts the interpretation given in the pioneer work of Shanabrook and Comas,[5] who assigned the dominant donor related line to the free heavy-hole–neutral-donor transition (HH-D^0). Though the assignment is the same as that proposed by the authors of Ref. 9, the spectra obtained presently differ significantly from those reported by these authors. In particular we do not find any trace of the two additional features assigned to the exciton bound to ionized donor (Si^+-X) and to the silicon–valence-band recombination. At least for the (Si^+-X) transition, this can be accounted for by the fact that the relative concentration of on-well-center Si^+ ions is much lower than in Ref. 9. Previous magnetotransmission experiments suggest that the Si^+ ions are indeed located near the barrier centers in our samples (Fig. 2 of Ref. 10). This is probably due to lower acceptor and "deep trap" concentrations in our samples as revealed by the lack of sensitivity of their magnetotransmission spectra to visible light illumination,[10,11] in contrast to the samples used in Ref. 9 (see also Refs. 6 and 7) which exhibited $1s\rightarrow 2p^+$ transitions of confined shallow (neutral) donors only after visible light exposure.

In the present investigations, we did not find any dramatic differences in the photoluminescence data collected for differently doped quantum wells (Fig. 4, Fig. 8). The same splitting of $E_B=1.8\pm 0.1$ meV between the X and the (D^0-X) lines is in very good agreement with the binding energy of an exciton bound to a neutral donor located at the center of the well as calculated by Kleinman.[18] Therefore, the impurity-related optical transitions in MQW are dominated by impurities located at the center of the well. This location corresponds indeed to the highest binding energy for such impurity centers, and to a singularity in the density of states.[1–4] Such a tendency, in the case of the (e-C^0) transition, has already been observed for intentionally undoped MQW,[25] in contradiction with the case of a single quantum well, where transitions were found to be related to acceptors near the interface.[28,29] This effect was explained by the large acceptor concentration near the AlGaAs/GaAs interface after growing a thick GaAlAs buffer.

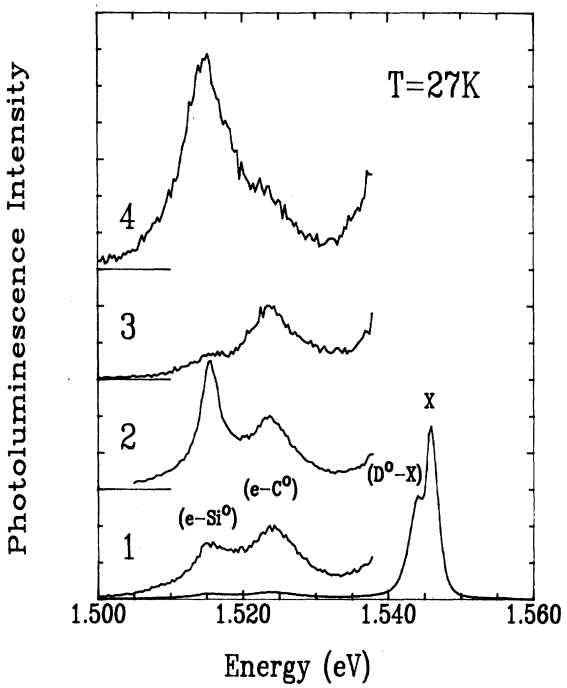

FIG. 8. A comparison of the acceptor-related photoluminescence spectra of samples 1–4, measured at 27 K. The spectra have been scaled and adjusted to the peak intensity of the free-electron–neutral-carbon-acceptor transition (e-C^0) in such a way that a direct comparison of th peak intensity of the (e-Si^0) transition can be made.

Far-infrared magnetotransmission experiments allow[10] the determination of the doping profile of the selectively doped MQW, provided that most of the impurities remain neutral, which is the case here. The results for samples 1–4 showed that the silicon donors are only in part located where they were intended to be. The same type of conclusion can be qualitatively obtained from the analysis of the luminescence experiments. This is clearly evidenced by the inspection of the free-electron–acceptor luminescence bands at 27 K for the different samples (Fig. 8). The carbon acceptors are nonintentional impurities which can be considered as homogeneously distributed in the sample. Since, as already pointed out, the luminescence peak corresponds to those acceptors locat-

ed at the center of the well, this (e-C^0) luminescence can be used as a local probe to compare the relative intensities of the (e-Si^0) transition and thus those of the relative concentration of silicon atoms at the center of the well. Care has to be taken, however, to work at a given temperature for all samples since the relative strengths of both transitions vary significantly with temperature (see Figs. 5 and 8 for instance). If we assume a constant level of residual C doping for all samples, one can see that the silicon related peak is, as expected, dominant for samples 2 and 4, in which intentional doping has indeed been realized in the center of the well. It is negligible for sample 3 but remains important for sample 1. All these results show that the incorporation of the impurities has been partly delayed by some kind of segregation process favoring the presence of donors in the center of the well for sample 1 but not by diffusion, which should favor the existence of these donors in sample 3 as well. Using the diffusion coefficients of atomic Si in GaAs as measured recently by Schubert *et al.*,[30] we can indeed confirm that impurity diffusion should not be very important in our case except maybe for the very first wells of our 150-period samples for which we evaluate the diffusion length to be 35 Å. A similar conclusion of the importance of segregation was obtained by Liu *et al.*[9] based on the analysis of luminescence features which are not observed in our samples. More quantitative information about the self-compensation can be obtained from the inspection of Fig. 8. Indeed, the concentration of residual C is of the order of 10^{14} cm^{-3}, that is, 10^8 cm^{-2} per well. The concentration of Si which incorporates in acceptor sites (As sites) is of the same order, that is, less than 5% of the total Si concentration. This result is interesting and shows that only a small fraction of the Si atoms remain in As sites at $T_s = 640$ °C despite the segregation process which leads them to migrate through several successive As planes. (The segregation length is estimated[10] to be of the order of 70 Å.)

The same analysis of the (D^0-X) transition at low temperatures (Fig. 4) is not so clear because we have no internal reference. However, when the spectrum of the sample 3 (doped at the end of the well) is compared to those of other samples, the main difference comes from the intensity of the (D^0-X) band significantly lower and the width of this transition significantly broader, reflecting the fact that transitions involving donors with smaller binding energies are now relatively important with respect to those related to the center of the well.

The magnetic-field dependence of the free exciton lines and acceptor-related transitions, analyzed in Sec. III (Fig. 2, Fig. 6), allows us to determine the binding energy of the carbon and silicon acceptors located at the center of the well. The values obtained are 29.7 ± 0.3 and 38.5 ± 0.2 meV for carbon and silicon acceptors, respectively. In the case of carbon, this value is slightly smaller ($\simeq 1$ meV) than that calculated by Masselnik *et al.*,[4] and experimentally evaluated by Miller *et al.*[25] However, this difference is essentially due to the different binding energies of the 1S heavy-hole exciton taken in the respective data.

The dominant character of the intrinsic transitions observed in quantum wells has been noticed in the pioneer work on 2D systems.[31] The peak intensity of the luminescence excitonic lines, measured at 4.2 K in δ-doped MQW, is two orders of magnitude higher than those observed in δ-doped bulk GaAs. This difference is well explained by the increase of the overlap of the hole and electron wave functions due to the bidimensionality.[31]

However, the comparison between results obtained for quasi-two-dimensional GaAs layers with those for bulk GaAs reveals that there is a difference between the relative intensity of excitonic transitions and donor-acceptor transitions. The experimental data obtained on samples 1–5 show clearly an enhancement of the process corresponding to the free exciton and to the exciton bound to a neutral donor and a disappearance of the donor-acceptor transitions, with respect to the same features observed in sample 6 (see Figs. 5 and 7). Most of the arguments invoked to explain these observations are dealing with the higher quality samples usually encountered with two-dimensional structures.[32] These arguments cannot be retained in our case since all our samples have been grown with the same technology and we do not have any evidence for accumulation of impurities at the interfaces of the MQW structures. Such possible accumulation of impurities at interfaces was ruled out by magneto-transmission experiments.[10]

We think that one of the main differences comes from the results concerning the statistics on random walks in 2D and 3D systems. The Polya theorem[33] states that: "In one or two dimensions two particles are certain to meet infinitely often, but in three dimensions there is a nonzero probability that they never meet." If we apply this theorem to the recombination of an electron-hole pair, we are led to think that the reduced dimensionality should favor the coupling of photocarriers into excitonic pairs which is manifested by the radiative efficiency enhancement of the related transitions. In the present study of intentionally doped samples these transitions are due to the recombination of free excitons and that of excitons bound to a neutral donor.

V. CONCLUSION

Photoluminescence experiments on differently δ-doped MQW were performed on samples already investigated by infrared magnetotransmission measurements.[10,11] The presence of Si acceptors has been detected for the first time in intentionally doped MQW. The analysis of the related luminescence features allows the confirmation that the planar doping procedure increases the efficiency of the location of impurities at the intended place. However, a noticeable amount of impurities drifts away. So photoluminescence experiments are found to be a very sensitive method of detecting impurity concentration in the central part of the well and to provide valuable information about the self-compensation and impurity segregation during the MBW growth of selectively doped MQW.

The comparison of the photoluminescence spectra of a selectively doped MQW and of a δ-doped GaAs bulk sample obtained by the same MBE technology shows that

the reduced dimensionality is the main reason for the radiative efficiency enhancement observed for excitonic transitions in quasi-2D systems.

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