PHYSICAL REVIEW B VOLUME 52, NUMBER 8 15 AUGUST 1995-II

Photoluminescence of the electron-dressed confined X^- exciton in an n-type AlAs/GaAs resonant tunneling device

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(Received 10 February 1995; revised manuscript received 22 March 1995)

Spectroscopic evidence is presented for the existence of the electron-dressed exciton (X^-)—an electron bound to an exciton—in the 8-nm-wide quantum well of an asymmetric n-type AlAs/GaAs triple-barrier resonant tunneling device. This is revealed by a transition just below the lowest heavy-hole exciton, leading to a doublet structure in the photoluminescence peak. The splitting of 2.1 meV is only weakly dependent on electrical bias. The effect is observed in the neighborhood of each of the three electron current resonances, when the number of electrons in the well is still low but significantly greater than that of photocreated holes. The influence on the X^- spectrum of applied electrical bias, temperature, excitation photon energy, and excitation power density is investigated. Indications were also found for the existence of X^- in the narrower, 6-nm-wide quantum well.

I. INTRODUCTION

Photoluminescence (PL) spectroscopy is a sensitive and effective technique for characterizing electronic and optical properties of resonant tunneling structures (RTS's) based on compound semiconductors,[1–11] which are commonly fabricated by molecular-beam epitaxy. Excitonic recombination inside the quantum well (QW) is a prominent feature in the PL spectrum of a RTS under bias. This results not only from the enhanced radiative recombination of confined excitons, but also from the efficient collection of carriers photocreated in the contact layers that subsequently tunnel into the QW.[2–5,8–11]

The existence of a bound state for different types of excitonic complexes was proposed by Lampert[12] in 1958 and further theoretical work has been performed on this subject.[13–15] In particular, a bound state was predicted for an electron-dressed exciton, denoted by X^-, composed of an electron bound to an exciton, with many similarities to the hydrogen anion H$^-$. The energy of the electron-dressed exciton is reduced compared to the sum of the separate free exciton and free electron due to the electron-electron exchange and correlation interactions, which is a genuine quantum physical effect. Spectroscopic evidence for X^- has been reported recently in three cases: in CdTe/CdZnTe multiple quantum-well systems,[16] in a bipolar resonant tunneling light-emitting diode,[17] and in modulation-doped GaAs/Al$_x$Ga$_{1-x}$As quantum wells,[18] an additional peak is observed in the optical spectra of the lowest heavy-hole exciton (denoted by X_{1h}). Also, a similar observation made recently in an n-type double-barrier RTS device, which was given a tentative explanation in terms of electron-hole exchange interactions,[19] should probably be ascribed to the same origin.

We present here PL results on the charged exciton X^- in an n-type AlAs/GaAs asymmetric triple-barrier RTS. By varying the applied bias across the device, the density of electrically injected electrons in the QW can be adjusted to create favorable circumstances for the X^- formation. With increasing density of the excess electrons in the QW's (i.e., electrons with density in the well in excess of that of the photocreated holes), the number of exciton-electron collisions increases and an exciton has a higher probability to couple to an electron, resulting in the X^- complex. The temperature evolution of the X^- component in the PL spectrum as well as the variations of its intensity with excitation photon energy (E_{exc}) and laser power (P) have been investigated.

II. EXPERIMENTAL PROCEDURES

The RTS used here is a unipolar device that is composed of two QW's, a 6-nm GaAs narrower well (NW) on top of an 8-nm GaAs wider well (WW) defined by three 4-nm AlAs barrier layers, all not intentionally doped. This intrinsic region is sandwiched between two 50-nm-thick n^--doped (5×10^{16} cm^{-3}) GaAs spacer layers, which in turn are surrounded by the GaAs top layer and the

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substrate, both n^+-doped (3×10^{18} cm^{-3}). Apart from its electrical effects, the use of n^--doped spacers tends to reduce the diffusion of donors into the intrinsic region during sample growth. The preparation of mesa and contact pad with an optical window of 30 μm in diameter for optical probing (similar to Ref. 11) allows measurements under electrical operation. Forward (reverse) bias designates applied positive (negative) voltage on the top electrode, i.e., the polarity for which the WW (NW) is the first well for electron tunneling. During PL measurements the device was mounted in a helium flow cryostat at temperatures down to 5 K. The laser power on the device was kept below 0.6 mW, at which no change is found in current-voltage (I-V) characteristics of the device. The optical path for PL studies was arranged in near backscattering geometry. Continuous wave optical excitation was performed by a Ti-sapphire laser, pumped by an Ar$^+$ ion laser. The laser spectrum was purified by a narrow-band prism filter. The light dispersion and multichannel detection of the emitted light were realized using a triple 80-cm monochromator and a nitrogen-cooled charge coupled device detector of high sensitivity, which yields a spectral resolution of 0.05 meV in the near infrared region. The size of the optical window of the device was deliberately chosen to match the excitation laser spot so as to maximize the signal-to-noise ratio.

III. RESULTS AND DISCUSSION

Figure 1 depicts the I-V characteristics of the device at $T = 5$ K, showing two current resonances in forward bias (1^+ and 2^+ at $V = +0.28$ and $+1.80$ V, respectively) and one resonance (1^- at $V = -1.20$ V) in reverse bias. In each case a so-called triple alignment occurs between the emitter and levels in the first and the second quantum well: E_1^{WW} and E_1^{NW} for the 1^+, E_2^{WW} and E_2^{NW} for the 2^+, and E_1^{NW} and E_2^{WW} for the 1^- resonance. Close to each of the three resonances, the accumulation of electrons in the two QW's can be significant, as observed in the same device by the intensity and width of the lowest exciton PL transitions in both wells.[20] One can see from Fig. 1 that at low voltage $-0.2 < V < +0.1$ V, the tunneling current is extremely small, as is the electron density in the QW's.

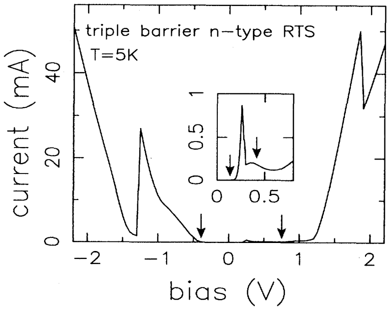

FIG. 1. Current-voltage characteristics of the triple-barrier RTS device measured at $T = 5$ K, showing the three tunneling current resonances in the voltage range of interest. The region of the narrow 1^+ resonance between the lowest levels in the two wells is enlarged in the inset. Arrows indicate the different regions where the doublets were observed.

The PL spectra in Fig. 2 were measured at a voltage close to the 1^+ resonance (see the inset of Fig. 1) in the region of the lowest exciton transition in the WW. They show the evolution of the lowest exciton PL peak with bias (a) before resonance and (b) after resonance. The PL transition at 1.576 eV in (a), appearing first at $V = +0.09$ V, stems from the radiative recombination of $n = 1$ free heavy-hole excitons (X_{1h}) in the WW. The X_{1h} excitons are mainly formed from photocreated electron-hole pairs because at this bias very few electrically injected electrons can tunnel into the WW. The broad feature at lower energy is extrinsic PL due to donors in the quantum well. It is described as (D^0, h) recombination with an estimated energy of 11 meV below the X_{1h} transition[21] for a donor at the center of the well.

Upon increase of the applied bias, the number of electrically injected electrons that tunnel into the WW increases and so does the electron density in the well. A new feature appears in the spectrum at the lower energy side of the X_{1h} peak, with a splitting in energy against the X_{1h} line of $\Delta E = 2.1$ meV. This feature grows very quickly with bias and dominates the PL band from $V = +0.11$ V on, over the whole 1^+ resonance region. One should be cautious about the latter statement: electron sheet densities above 10^{11} cm^{-2} can easily occur in the resonance region changing the character of the transition, because above this density screening effects can inhibit the formation of excitons.[15,22]

The WW-to-NW alignment exists only for a narrow bias region, yielding a very sharp feature in the I-V characteristics at the 1^+ resonance (inset of Fig. 1). Beyond the resonance, the X_{1h} line becomes observable again at $+0.36$ V as shown in Fig. 2(b). Since the alignment between the WW and the emitter is still locked in this bias range, the lower-energy feature does not disappear until

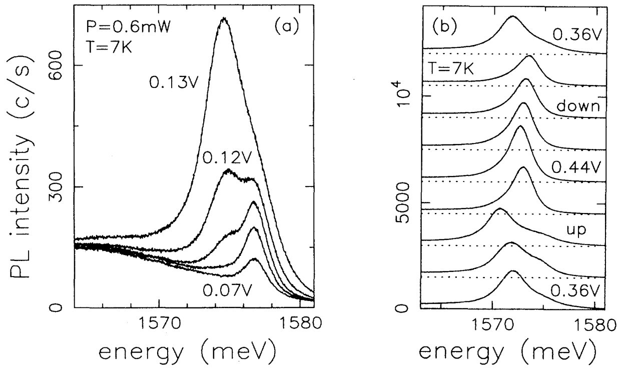

FIG. 2. Evolution of the doublet structure in the WW PL spectra ($E_{\mathrm{exc}} = 1.658$ eV) taken when the external bias is swept over the 1^+ resonance, (a) below the resonance and (b) above the resonance, at $T = 7$ K for increasing and for decreasing voltage (steps of 20 mV). Also shown in (b) is the hysteresis in the line shape for discharging ("up" scan) compared to recharging ("down" scan) of the wide QW.

the density of excess electrons in the WW decreases to such a extent that the X_{1h} line regains dominance over the feature. This happens suddenly at $V = +0.42$ V [between the third and fourth curves in Fig. 2(b)], which points to a sudden disruption of the emitter-to-WW alignment.

The close correlation between the existence of the lower-energy feature and the density of excess electrons in the QW indicates that this feature must be related to an excitonic complex of larger binding energy ΔE than the excitons, which is nothing but the electron-dressed exciton X^-. From Ref. 23, and taking recent material parameters valid for GaAs at $T = 5$ K, a theoretical value of $\Delta E = 2.2$ meV is found for X^- relative to X_{1h} in a two-dimensional system, in excellent agreement with the observed splitting. The evolution of the spectra in Fig. 2(a) is very similar to that reported by Langerak *et al.*,[19] which was ascribed to electron-hole exchange splitting, but in our opinion also originates from the X^- exciton.

Doublet structures in the emission spectra of confined excitons in QW's have been reported in several instances and ascribed to a variety of origins. We briefly consider the proposals different from the X^- formation in connection with the present results. Excitons bound to acceptors or donors can possess a binding energy comparable to the doublet splitting observed here.[24–26] However, the saturation properties of the present PL peak do not agree with extrinsic PL (see below). The biexciton[27,28] possesses a binding energy comparable to that of X^-, but its formation is expected in conditions of high exciton concentration and will be favored by balanced electron and hole densities, both of which are not the case here. Discrete well-width fluctuations[29] seem to be excluded because they would show up more strongly in the NW than in the WW spectra. Finally, the electron-hole exchange effect[30–32] is sometimes invoked, which can cause an increase in exciton binding energy. This cannot be the origin of the doublet splitting observed in our PL measurement because the electron-hole exchange interaction in the NW should again be considerably larger than in the WW due to larger overlap of the wave functions. Moreover, regardless of which of the alternative origins—donor or acceptor, biexciton, well-width fluctuations, or exchange splitting—it is hard to give a good explanation for the bias dependence of the spectrum reported here. The X^- exciton yields a natural explanation for the appearance of the emission line each time the electron density is increasing over a certain threshold.

Since the existence of the X^- line closely depends on the concentration of excess electrons in the QW, one would expect it to be sensitive to the space-charge effect,[33] which can occur at resonance and causes hysteresis effects in the device operation. This is exactly what is observed in Fig. 2(b), showing spectra recorded beyond the resonance with increasing and decreasing bias variation. A hysteresis of about 40 mV in applied voltage is clearly seen by comparing the corresponding spectra, paying special attention to the disappearance and reappearance of the doublet structure. The hysteresis is a direct reflection of space-charge buildup in the QW when it proceeds in discharging ("up" scan at $V = +0.42$ V) and recharging ("down" scan at $V = +0.36$ V) close to the current resonance. The behavior of the spectra in Fig. 2(b) is a different manifestation of the fact that a sufficient density of excess electrons in the QW is a prerequisite for the X^- formation.

The influence of temperature on the doublet structure is shown in Fig. 3(a), where the X^- intensity gradually decreases with respect to X_{1h} when the temperature increases. In good agreement with the observed binding energy ($\Delta E = 2.1$ meV) the X^- line nearly vanishes when temperature goes up to 30 K. This results from thermally induced dissociation of X^- into the free exciton and the free electron. Figure 3(b) shows, on a logarithmic scale, the integrated intensity ratio between the two lines as a function of the inverse temperature, which clearly demonstrates the thermal activation with an energy difference of 2.1 meV, corresponding to the measured doublet splitting. The intensity ratios are obtained from a profile analysis using two Voigt components (convolution of Gauss and Lorentz) and a linear base line.

The X_{1h} component evolves from a purely Gaussian shape at $T = 5$ K with a linewidth of only 1.8 meV to a more Lorentzian one at high temperature. Such a narrow linewidth is found because the PL originates from a single quantum well (no fluctuations between subsequent wells), the electric field is low and therefore so are the field inhomogeneities, and the interface quality is very good. From the line-shape analysis, the X^- line has a Lorentzian fraction comparable to the Gaussian one already at low temperature. At $T = 35$ K both lines are predominantly Lorentzian with full width at half maximum (FWHM) of 2.3 meV and 3.6 meV for X_{1h} and X^-, respectively.

The low-temperature linewidths clearly result from inhomogeneous broadening. The appreciable temperature dependence of the line shapes and widths can have different origins. We can exclude interactions with the LO

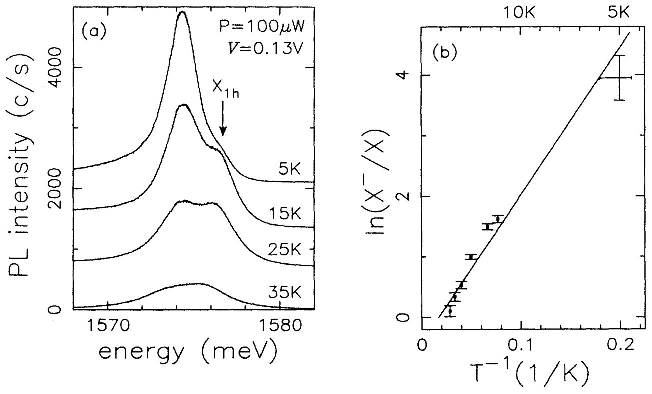

FIG. 3. (a) PL spectra of the WW measured as a function of temperature taken before the 1^+ resonance at $V = +0.13$ V. (b) Ratio of integrated intensities of the X^- to X_{1h} band on a logarithmic scale as a function of inverse temperature. The solid line is a guide to the eye, the slope of which corresponds to the thermal activation energy of 2.1 meV.

phonons at these low temperatures. Also the tunneling escape times are not expected to be changing in this temperature range. The scattering by acoustic phonons was shown for the normal exciton to yield a contribution to the FWHM smaller than 0.5 meV (Ref. 34) below $T = 35$ K, well below the measured widths (after scaling for the smaller well width of our sample). Because of fluctations in the quantum-well potential, e.g., due to atomic steps of the interfaces, the exciton is localized at low temperature. At higher temperature it can reach regions of somewhat higher potential, corresponding to a different transition energy, which can be an origin of the temperature dependent line shape. Finally, it is not excluded that carrier-carrier interactions lead to a homogeneous broadening of the X_{1h} and X^- transitions, taking into account the excess electrons that are electrically injected in the QW.

It should be stressed that the intensity ratio X^-/X_{1h} also depends on the bias voltage due to the different amounts of excess electrons, but the thermal activation energy is expected to be the same. The X^- line intensity is transferred to the X_{1h} line, while the overall intensity is decreasing. The latter results from the decrease in recombination efficiency for QW emission at higher temperatures.[35,36]

In addition to the observation of the doublet structure around the 1^+ resonance, we have also found similar effects in a narrow bias range before the 2^+ resonance in forward bias and before the 1^- resonance in reverse bias, as shown in Fig. 4. Compared to Fig. 2, the transition energy of both X^- and X_{1h} is much lower, especially in the case before the 2^+ resonance [Fig. 4(a)], which is caused by the quantum confined Stark effect at high bias. In addition, the doublet splitting is not as sharp as that observed near the 1^+ resonance, corresponding to slightly wider linewidths of the components. The latter could be due partly to static effects, e.g., larger lateral inhomogeneity of the electric field over the device at higher absolute bias voltage, but also to dynamic effects, which reduce the dephasing time. The splittings determined from the profile analysis are $\Delta E = 2.1$ and 2.4 meV before the 2^+ and 1^- resonances, respectively. These small differences in ΔE are not understood in detail.

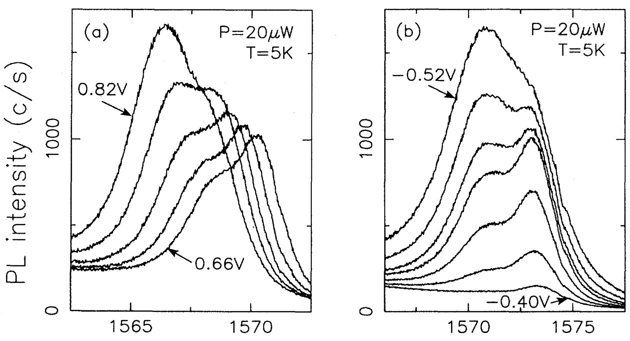

FIG. 4. Appearance of the doublet structure in the WW emission ($E_{\rm exc} = 1.687$ eV) measured at $T = 5$ K when the bias voltage is approaching (a) the second forward (2^+) and (b) the reverse bias (1^-) resonance. The voltage scan step is (a) 40 mV and (b) 20 mV.

The spectra in Fig. 5(a) are obtained at increasing $E_{\rm exc}$ using the same laser power density $P = 14$ W/cm^2. These energies correspond to direct optical excitation in the GaAs contact layers only ($E_{\rm exc} = 1.558$ eV), in the contact layers and the WW (1.610 eV), and in all GaAs layers including the WW and the NW (1.687 eV), since in this device the transition energies of the lowest free heavy-hole excitons in the WW and in the NW at the +1 resonance are 1.575 eV and 1.650 eV, respectively. The increasing hole density in the WW with increasing photon energy yields an appreciable enhancement (up to a factor 3) of both X_{1h} and X^- peaks. This is not visible here because the spectra are normalized to the X_{1h} component in order to clearly demonstrate the relative increase in intensity of X^- with respect to X_{1h}. Indeed, for the lowest energy case (1.558 eV) electron-hole pairs are only created in the GaAs contact layers and the only source of holes is the accumulation layer on the collector side, from which they can tunnel through the barriers and the NW into the WW. For the direct optical excitation in the NW, there are three hole sources available for the WW, i.e., the WW itself, the NW, and the hole accumulation layer on the collector side. However, the small but consistent increase of the X^- intensity relative to that of the X_{1h} shown in the figure is ascribed to the increased number of photoexcited electrons. Indeed, the X^- and X_{1h} densities are in a first approximation expected to be quadratically and linearly dependent, respectively, on the electron density in the QW. The electron density (electrically injected as well as photocreated) should increase by 5–10 % in order to explain the change in relative intensities observed in the figure. This is acceptable, regarding the very low tunneling current at the measuring voltage, which reflects in a low density of electrically injected electrons.

Using excitation at 1.687 eV, Fig. 5(b) shows the dependence of the spectrum on the laser power on the device, ranging between 14 W/cm^2 and 84 W/cm^2. No

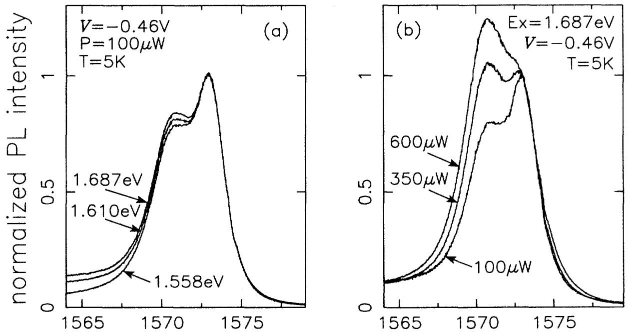

FIG. 5. PL measurements of the WW emission before the 1^- resonance, normalized to the X_{1h} exciton intensity, and measured (a) for different excitation photon energies $E_{\rm exc} = 1.558$, 1.610, and 1.687 eV at a constant power density $P = 14$ W/cm^2, and (b) for increasing $P = 14$, 49, and 85 W/cm^2 at $E_{\rm exc} = 1.687$ eV.

saturation at all is found for the intensity of the X^- line even at higher excitation powers, in contrast to the case of impurity-related bound exciton emission, which can be easily saturated[26,32] at levels as low as 10 W/cm^2. This definitely rules out models in which the lower-energy component in spectral doublet splitting would be assigned to extrinsic exciton transitions. The similarity of the spectra in Figs. 5(a) and 5(b) shows once more that the growth in intensity of the X^- line with the number of photoexcited carriers at constant excess electron densities is faster than for X_{1h}. When increasing the excitation power, we increase the number of photocreated electrons and holes in much the same way as by variation of E_{exc}.

In our measurements, no clear doublet splitting could be observed in the NW PL spectra around any of the three current resonances. However, indications for unresolved splitting are found at the 1^+ resonance, where the electron accumulation in the well is significant. Figure 6 presents the PL spectra of the NW when applied bias is swept through the 1^+ resonance: Before and after the resonance the PL band is more symmetric and at the resonance ($V = +0.28$ V) a bump appears at the low-energy side. A profile analysis, which unfortunately does not yield a unique solution, is able to give a very good fit, taking one Gaussian component with similar width and position as the symmetric NW peaks (with a FWHM approximately equal to 4 meV) and one Voigt component at lower energy by about 2.6 meV (see dotted lines in Fig. 6). In this picture, the X^- exciton would only exist exactly at the resonance, when a sufficient density of excess electrons is built up. The splitting is not resolved only due to the somewhat larger linewidths than for the WW transitions.

In the same figure we also depict the asymmetric line shape occurring in reverse bias in a limited range around -0.45 V, much earlier than the 1^- resonance. This is very similar to the lineshape at $V = +0.28$ V and invites the same interpretation as given above. It is not so clear why in this region the population of X^- excitons would go through a maximum. Indeed, higher electron densities in the well are expected in the NW when one gets closer to the resonance, and at first sight this would be more favorable for X^- formation.

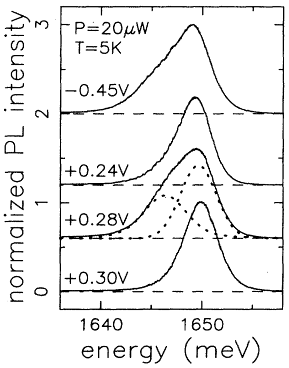

FIG. 6. Asymmetric PL peak of the narrow QW at the 1^+ resonance ($V = +0.28$ V) compared to more symmetric lines at nearby bias values. The dotted line shows a decomposition of the PL peak in one Gaussian and one Voigt component. A similar asymmetry of the PL spectrum is found in reverse bias ($V = -0.45$ V) before the 1^- resonance.

It is worth pointing out that previous experimental observations were also made on relatively wide wells: Two of the reports about the X^- doublet spectra in GaAs pertain to a well width of 9 nm, comparable to that of the WW in our sample, and nearly the same binding energies were measured [$\Delta E = 2$ meV (Ref. 17) and 1.9 meV (Ref. 19)]. In the third case, for a modulation-doped GaAs/Al$_x$Ga$_{1-x}$As system,[18] a splitting of only $\Delta E = 1.2$ meV is measured, which could be a result of the very different type of confinement potential. Taking into account the smaller effective size of excitons[37] in CdTe, the 10-nm well of Ref. 16 is equivalent to a well even wider than a 9-nm GaAs well.

IV. CONCLUSIONS

In an n-type AlAs/GaAs asymmetric triple-barrier RTS, the density of electrons in the quantum wells can be adjusted by the external bias that controls the electron tunneling processes. In the presence of excess electrons in the 8-nm-wide QW, the $n = 1$ free heavy-hole exciton has a higher probability to couple to an electron through electron-electron exchange and correlation interactions, forming an electron-dressed exciton X^-. The low-energy component of the doublet structure, which we have observed in the low-temperature PL spectra of the WW, is ascribed to the recombination luminescence of this electron-dressed exciton. The binding energy of the X^- exciton is found to be $\Delta E = 2.1$ meV for an 8-nm-wide GaAs quantum well. The electron-dressed exciton thermally dissociates to a free exciton and an independent electron, with ΔE the activation energy. It is clearly demonstrated that the excess electron density in the quantum well is a decisive factor for the formation of the X^- emission line.

In the narrower (6 nm) QW the X^- peak is not resolved. At the 1^+ resonance and in reverse bias well below the resonance, a feature is observed at the low-energy side of the PL peak that possibly originates from the X^- exciton in this well.

ACKNOWLEDGMENTS

Several interesting discussions with F. Peeters are gratefully acknowledged. We thank the Belgian National Fund for Scientific Research and the Inter-University Institute for Nuclear Physics for financial support. This work was also supported by a cooperation program of the Inter-university Microelectronics Center and the Flemish universities. Z.C.Y. wishes to thank the Antwerp province for support given in the framework of the exchange program between the sister provinces Antwerp and Shaanxi.

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