Resonant tunneling and photoluminescence spectroscopy in quantum wells containing self-assembled quantum dots


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We investigate the optical and electrical properties of $n$-$i$-$n$ GaAs/(AlGa)As double barrier resonant tunneling diodes (RTDs) in which a layer of InAs self-assembled quantum dots (QDs) is embedded in the center of the GaAs quantum well. A combination of photoluminescence (PL) and electrical measurements indicates that the electronic states and charge distribution in this type of RTD are strongly affected by the presence of the dots. Also, the dot PL properties depend strongly on bias, being affected by tunneling of majority (electrons) and minority (photocreated holes) carriers through the well. The measurements demonstrate nonlinear effects in the QD PL by means of resonant tunneling and the possibility of using the dot PL as a probe of carrier dynamics in RTDs.

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I. INTRODUCTION

Since the first experimental observation of resonant tunneling, there has been a great deal of interest in the physics of this phenomenon and its potential application to high speed electronics and optoelectronics. Optical spectroscopy on $n$-$i$-$n$ resonant tunneling diodes (RTDs) reveals resonant changes in the quantum well luminescence as a function of applied bias. This luminescence arises from the recombination of electrons (majority carriers) from the doped emitter layer with minority holes, created either by light or by impact ionization, which tunnel into the quantum well from the opposite side of the device. Efficient electroluminescent RTDs can be obtained by sandwiching the double barrier structure in the intrinsic ($i$) region of a forward-biased $p$-$i$-$n$ diode. These devices have been proposed as potential fast optical modulators. Also, it has been demonstrated that resonant tunneling can be used as an efficient method of injecting carriers into the active region of a semiconductor laser diode.

In this article, we investigate resonant tunneling and radiative recombination of electrons and holes in a series of $n$-$i$-$n$ GaAs/(AlGa)As double barrier RTDs in which a layer of InAs self-assembled quantum dots (QDs) is embedded in the center of the GaAs quantum well (QW). Resonant tunneling in quantum dot structures has been reported previously. The dots were either incorporated in a single AlAs tunnel barrier or else in, or close to, a GaAs quantum well surrounded by two (AlGa)As barriers. Tunneling spectroscopy has been used to study transport through the zero-dimensional electronic states of a dot and to realize QD-based memory devices. However, to date little information exists on the carrier dynamics and optical properties of tunneling devices in which the quantum well contains QDs, a system of relevance to the operation of QD lasers and other devices incorporating QDs.

The article is organized as follows. In Sec. II we describe the device structure. Section III describes the electrical and optical properties of the devices. We show that the insertion of the InAs layer has a profound effect on the QW electronic structure, strongly modifying the QW quasibound states with even parity (i.e., those with a maximum probability density at the center of the well, where the InAs layer is located). This results in a marked change of the current–and capacitance–voltage characteristics of the RTDs compared to a control sample with no dots in the QW. In addition to the modification of the electronic structure, the presence of the QDs also induces charge redistribution inside the RTD. We show that at zero bias dots are filled with electrons. The associated negative charge accumulation induces the formation of depletion regions adjacent to the (AlGa)As barriers, thus shifting the electron subbands of the QW to higher energies with respect to a sample containing no dots. In Secs. IV and V, we investigate carrier tunneling and charge distributions in and around the QW when the device is excited by laser light. A strong resonance in the photocurrent characteristics is observed and attributed to variations in the number of holes tunneling into the well. Magnetotunneling spectroscopy is used to confirm this attribution (Sec. IV). The same resonance together with other similar additional resonances (not visible in the current–voltage characteristics under illumination) are observed as maxima in the dot photoluminescence (PL) intensity with varying bias (Sec. V). These origi-
nate from recombination of majority (injected electrons) and minority (photogenerated holes) carriers which are photogenerated in, and tunnel from, the depletion layer of the RTD. The voltage-tunable resonant changes in the dot luminescence are discussed in terms of the tunneling times and of the capture times of carriers into the dots.

II. DEVICE STRUCTURE

The RTDs were grown by molecular beam epitaxy on (100)-oriented GaAs substrates. Structures $q1d$ and $q2d$ consist of two 8.3-nm $Al_{0.3}Ga_{0.7}As$ barriers and a 12-nm GaAs QW in the undoped intrinsic ($i$) active region of an $n$-$i$-$n$ structure. A layer of InAs dots having different dot size and density was grown in the center of the well by depositing 1.8 (sample $q1d$) and 2.3 (sample $q2d$) monolayers (ML) of InAs. Undoped GaAs spacer layers of width 50 nm separate the $Al_{0.3}Ga_{0.7}As$ barriers from $2 \times 10^{17}$ cm$^{-3}$ $n$-doped GaAs layers of width 50 nm. Finally, $3 \times 10^{18}$ cm$^{-3}$ $n$-doped GaAs layers of width 0.3 $\mu$m were used to form contacts. A schematic diagram of the device is shown in Fig. 1. Also a wetting layer (WL) sample (sample $wl$) was obtained by growing the InAs layer on a (311)B GaAs substrate, keeping the same growth scheme of sample $q1d$. Finally, for comparison purposes, a control sample $c$ was grown with the same sequence of layers but with no InAs layer. These structures were grown at 600 °C except for the InAs layer and the overgrown 5.6-nm GaAs barrier, which were grown at 480 °C to avoid In segregation and desorption effects.

Each sample was processed into circular mesa structures of diameter 100 or 200 $\mu$m. A ring-shaped electrical contact was fabricated on the top of the mesa to permit optical access to the sample for measurements of PL and the current–voltage [$I(V)$] characteristics under illumination. The optical excitation was provided by the 633-nm line of a He–Ne laser or the 515-nm line of an Ar$^+$ laser. $I(V)$ and capacitance–voltage [$C(V)$] measurements were made on the same mesas used for the optical measurements. The $C(V)$ measurements were made with an HP 4275A impedance meter over the frequency range 10 kHz–1 MHz.

III. RTD ELECTRICAL AND OPTICAL PROPERTIES

Figure 2 shows the PL spectra at 10 K for all samples at zero bias. The PL of sample $wl$ shows a narrow band at $\sim 1.4$ eV due to carrier recombination in a two-dimensional InAs layer (wetting layer, WL). The PL of sample $q1d$ ($L = 1.8$ ML) and $q2d$ ($L = 2.3$ ML) shows a broad band (QD) at 1.35 and 1.25 eV, respectively, due to carrier recombination in the dots. The redshift of the QD band with increasing $L$ is a consequence of the increase in size of the dots. Finally, the PL spectrum of the control sample, $c$ (see the inset of Fig. 2) shows bands QW and A, which originate from recombination of carriers in the lowest electron and heavy-hole states of the GaAs well and in the GaAs doped layers, respectively. The band QW is absent in samples $wl$, $q1d$, and $q2d$. The spectra indicate clearly that the InAs layer introduces optically active electronic states below the GaAs conduction band edge, which efficiently capture the carriers generated in the QW. As we will see in the following, this has profound consequences for the $I(V)$ characteristics.

Figure 3 shows the $I(V)$ characteristics of the four devices in forward bias (negatively biased substrate). They differ substantially from each other: In the control sample two resonances, $e1$ and $e2$, are observed corresponding to electrons tunneling through the first two quasibound states of the QW; in contrast, in samples $wl$, $q1d$, and $q2d$, the low bias resonance is not observed and the second resonant peak is shifted to higher voltages. In addition, in the case of the $wl$ sample, we observe a peak in the differential conductance $dI/dV$ around zero bias [see the inset of Fig. 3 (b)].

As a preliminary to analyzing the optical and electrical properties of RTDs incorporating an InAs layer, we first modeled the electronic levels of this type of structure. Figure 4 shows a schematic diagram of the electron eigenstates of a GaAs/$Al_{0.3}Ga_{0.7}As$ QW with and without a uniform InAs layer. We solved the Schroedinger equation for the system in
the effective mass approximation for a GaAs well having a width equal to the nominal value (12 nm) and incorporating a two-dimensional InAs layer (i.e., no QDs) of thickness $L = 1.5$ ML. The effective mass theory is a reasonable approximation in the case of a very thin InAs layer because the wave functions extend into the confining barrier layer.\(^{21}\) The value of the electron effective mass, $m^*\approx m_e$, in the InAs layer is assumed to be equal to that of GaAs ($m^* = 0.067 m_e$, where $m_e$ is the free-electron mass). Although the band-edge effective mass of bulk InAs is small ($m^* = 0.023 m_e$), a substantially higher value is more appropriate to our model due to the nonparabolicity of the conduction band. The assumptions on which our model is based are justified in the limit of these thin InAs layers, since the calculation is mainly sensitive to the barrier parameters. Our model takes into account the strained gap of InAs\(^{22}\) and uses conduction band offsets of 700 and 400 meV for the InAs/GaAs and the GaAs/AlGaAs well, respectively. The calculation was repeated for different $L$ in the range 1–2 ML and did not show any qualitative difference from that for $L = 1.5$ ML. The effect of the thin InAs layer is to lower the energy of the QW ground state ($e1$) below the GaAs conduction band edge. In contrast, higher energy states are less strongly perturbed, particularly the odd-parity states having nodes at the center of the well.\(^{23}\) Our model shows that the InAs layer lowers $e1$ below the GaAs band edge, thus making $e1$ unavailable for resonant tunneling, consistent with the observed absence of the $e1$ resonance in the $I(V)$ of RTDs containing the InAs WL (sample $wl$). Similarly, the PL transition from $e1$ to the valence band, observed in the control sample (no InAs layer), is absent in the WL structure. The above model represents an oversimplified picture for samples $qd1$ and $qd2$. In fact, it neglects the real morphology of the InAs layer, which consists of dots and a two-dimensional InAs WL formed beneath them.\(^{17}\) However, since the dots can be represented as local minima in the WL potential profile, we can expect that the features related to $e1$ will be absent in both the $I(V)$ curves and the PL spectra of samples $qd1$ and $qd2$.

The modification of the electronic levels of the RTD due to the presence of the InAs layer induces a redistribution of the electron charge in the device and leads to major changes in the electrostatic potential profile. Due to the presence of InAs levels below the GaAs conduction band, at zero bias some electrons diffuse from the doped GaAs layers into the InAs layer states. This effect is monitored by capacitance–voltage measurements. $C(V)$ curves at $T = 5$ K are shown in Fig. 5 for samples $c$, $qd1$, and $qd2$. The capacitance is defined as $C = dQ/dV$, where $dQ = dQ_{acc} + dQ_{QW} = -dQ_{dep}$ is the incremental change of the negative charge in the accumulation layer on the emitter side ($dQ_{acc}$) and in the QW ($dQ_{QW}$), for an incremental change in the applied voltage $dV$.\(^{24-27}\) The corresponding increase in the positive charge in the depletion layer is $dQ_{dep}$. In all samples we observe an increase of capacitance when the bias is large enough to form an electron accumulation layer adjacent to the (AlGa)As barriers. In sample $c$, the capacitance rises at a relatively low bias since an accumulation layer quickly forms beneath the GaAs band edge ($V_{th}$) (see Fig. 5) in the $C(V)$ curves of samples $qd1$ and $qd2$ can be explained by the fact that at zero bias, electrons accumulate in the dots and depletion regions form in the region beyond the (AlGa)As barriers as
shown in the inset of Fig. 5. Consequently, a voltage $V_{th}$ is required to reach flat-band conditions and to form an electron accumulation layer adjacent to one of the barriers. This also has the effect of raising the energy of the electron levels of the well relative to the conduction band edge in the doped GaAs contact layer. Consistent with this, we observe a shift in the voltage position of the $e2$ current peak in samples $qd1$ and $qd2$. $V_{th}$ indicates the threshold voltage for the increase of capacitance in samples $qd1$ and $qd2$. The inset shows a sketch of the RTD potential profile at zero bias for the control sample (c) and a RTD containing dots (qd).

**IV. CURRENT–VOLTAGE CHARACTERISTICS UNDER ILLUMINATION**

We now consider the effect of optical excitation on the $I(V)$ characteristics of the devices. Fig. 6 illustrates the dynamics of carriers when the device is excited with above-band gap laser light. Electrons are electrically injected from the negatively biased GaAs emitter layer into the well. At the same time, light creates photocarriers (electron–hole pairs) throughout the sample. The electric field in the depletion layer separates the oppositely charged carriers: The photoelectrons are swept into the positive contact (electron collector), whereas the holes are attracted by the negatively biased electron emitter and form an accumulation layer adjacent to the right-hand barrier. During resonant transmission of electrons and photogenerated holes through the well, some of the carriers can be captured by the dots and then recombine radiatively. Carrier capture into QDs proceeds within times $\tau_c \sim 1 \text{ ps}$, much shorter than the characteristic dwell times ($\tau_d$) of electrons and holes that tunnel resonantly into the well, but comparable to, or even longer than the time ($\tau_I$) for carriers to make a single (semiclassical) transit of the QW and barrier region. Due to this fast carrier capture, the bias dependence of the dot luminescence is thus very sensitive to the resonant tunneling process.

The $I(V)$ characteristics under illumination for sample $qd2$ ($L = 2.3 \text{ ML}$) are shown in Fig. 7. In the presence of illumination, the current initially increases with bias and then reaches an almost voltage-independent saturation level that depends linearly on the laser excitation power. In addition, a resonant current peak, $h^*$, appears in $I(V)$ recorded at high illumination levels.
According to the carrier dynamics depicted in Fig. 6, the photocurrent depends on the carrier generation rate in the depletion region. It increases with the incident optical power and with the thickness of the depletion region, τ, and it should be relatively insensitive to whether or not the holes accumulated against the right-hand barrier are in resonance with the QW hole resonant states, i.e., they will eventually tunnel towards the QW and electron emitter, whether the process is resonant or not. However, their density in the hole accumulation layer is sensitive to their tunneling rate and will affect the electrostatic profile beyond the second barrier, giving an electrostatic feedback effect,\textsuperscript{30} as discussed in detail below. The initial increase of photocurrent with bias is due to the increasing extent of the depletion region, where the screening charge is due to ionized donors ($\sim 10^{15}$ cm$^{-3}$) in the GaAs depletion layer. This increase saturates when the depletion region extends into the heavily doped ($2 \times 10^{17}$ cm$^{-3}$) GaAs regions ($t \approx 50$ nm). The subsequent increase of the depletion width with bias is very slow, resulting in the saturation of current for $V>\sim 0.2$ V. We believe that the presence of the resonance $h'$ in $I(V)$ reflects resonant tunneling of the photocreated holes from the hole accumulation layer into the well. At the $h'$ resonance, the holes can readily tunnel into the well. This reduces the number of holes adjacent to the right-hand barrier. The resulting decrease in hole screening increases the width of the depletion layer and, through an electrostatic feedback effect, increases the rate of hole photogeneration. At higher bias, the depletion layer thickness is almost constant, so the resonant feedback effect is absent. This accounts for the absence of additional resonances in $I(V)$. Finally, the resonance $h'$ can be observed only at the higher illumination level because only in this case does the current remain unsaturated in the bias range of the $h'$ resonance.

In order to study further the origin of the $h'$ resonance, we examine its dependence on magnetic field. Magnetotunneling spectroscopy has proved to be a powerful means of distinguishing electron and hole tunneling and of measuring hole dispersion curves in a QW.\textsuperscript{31,32} Figure 8(a) shows the $e2$ resonant current peak (measured without illumination) for different magnetic fields, $B$, applied in the plane of the tunnel barriers in sample $qd2$. A similar plot is shown in Fig. 8(b) for the $h'$ resonance measured under illumination in the same sample $qd2$. Both resonances shift towards high voltages with $B$. However, the magnitude of this shift is different [see the inset of Fig. 8(a)] and can be accounted by the different effective masses of carriers involved in the tunneling.

For a parabolic band of effective mass $m^*$, the shift in bias, $\Delta V$, of the resonances can be written as

$$e \Delta V = f \left( \frac{eB\Delta s}{2m^*} \right)^2,$$

where $f$ is the ratio between the total voltage and the potential difference between the accumulation layer and the center of the well, and $\Delta s$ is the total distance traveled by carriers.
tunneling out of the accumulation layer into the QW. According to Eq. (1), the ratio of the shifts for the $e_2$ and $h'$ resonance is given by

$$\frac{f_e \Delta s_e m_e^*}{f_h \Delta s_h m_h^*}.$$  

(2)

By using an electrostatic model for the device, we estimate that $f_{h'} \sim 2$ at $h'$ ($V \sim 0.2$ V, under illumination) and $f_{e} \sim 4$ at $e_2$ ($V \sim 1.0$ V). Using the Fang–Howard approximation, we obtain $\Delta s_e \sim 28$ nm and $\Delta s_h \sim 22$ nm. The experimental value $r = 6.2$ ($= \Delta V_f/\Delta V_h$) therefore indicates that the ratio of the in-plane effective masses is $m_{e}^*/m_{e} \sim 2.4$. Given that $m_{e}^* = 0.067 m_e$ (where $m_e$ is the free-electron mass), we obtain $m_{h}^* \sim 0.17 m_e$. This is smaller than the bulk heavy-hole mass of GaAs ($0.45 - 0.57 m_e$), but this is not unexpected since the in-plane effective mass for higher energy hole subbands in GaAs/(AlGa)As QWs can be smaller. In particular, according to the simple model presented in Sec. III, the first level available for hole resonant tunneling is the second QW heavy hole state and a value for $m_{h}^*$ intermediate between the light-hole and the heavy-hole mass is expected. These measurements give a rough indication that our assignment of $h'$ to resonant tunneling of holes is correct.

The $I(V)$ characteristics clearly demonstrate the important role of minority carriers in determining the electrical properties of our RTDs under illumination. In the next section, we will show how the combination of optical and electrical excitation can be used to induce an electrically controlled change of the PL spectrum of the QDs.

V. BIAS DEPENDENCE OF THE QD PHOTOLUMINESCENCE

When a bias is applied, we observe significant changes in the luminescence from the quantum dots: the intensity, energy position, and linewidth of the QD PL are all sensitive to bias. Figure 9 shows the voltage dependence of the peak intensity of the dot photoluminescence $PL_{QD}$ at 10 K for different light excitation levels $P$ in sample $qd2$ ($L = 2.3$ ML). With increasing $V$, the dot PL intensity increases sharply and shows resonant peaks labeled $h'$, $h''$, and $h'''$. The diode is excited with above-band gap laser light (633 nm) at different laser power densities, $P (P_0 = 3$ W/cm$^2$). The inset shows a comparison between the voltage dependence of $PL_{QD}$ (continuous line) and the peak energy position of the dot PL band, $h \nu_{QD}$ (full dots) for $P = 9 P_0$. $\sim 10^{-7} - 10^{-5}$ is the transmission coefficient of the tunnel barriers estimated from the device parameters. The long dwell time corresponds to a resonant increase in the probability density of the hole wave function in the well. A carrier capture time of 1 ps (Ref. 28) corresponds to a hole cross section for single dot of $\sigma_{QD} \sim 10^{-12}$ cm$^2$, so that the probability of hole capture cross section on a single transit time is small, $\sigma_{QD} N_{QD} \leq 0.1$. Therefore, off resonance, most of the holes make a single pass of the quantum well without interacting with the QDs; they pass through both barriers and recombine with electrons in the electron emitter region. In contrast, on resonance the dwell time in the QW is sufficiently long for almost all of the resonant holes to be captured onto the dots, where they recombine with the electrons on charged dots. This gives rise to a resonantly enhanced $PL_{QD}(V)$ (see resonances $h'$, $h''$, and $h'''$ in Fig. 9). This description is confirmed by the observation of the weak antiresonant structure in the PL intensity $PL_{A}$ of the band A due to carrier recombination in the bulk GaAs layers. The bias dependence of $PL_{A}$ is plotted in Fig. 10 and shows an antiresonant behavior with respect to that of $PL_{QD}$, i.e., the intensity of A has a minimum at the maximum of resonance $h'$, corresponding to a reduced number of holes recombing in the GaAs emitter layer due to the efficient hole capture into the dots.

These measurements indicate that the hole capture into the dots and the subsequent recombination with electrons...
changes in the dot photoluminescence with bias and as means of probing the tunneling process.

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VI. CONCLUSION

We have investigated the influence of InAs QDs on the electrical and optical properties of resonant tunneling diodes in which the dots are embedded in the center of the GaAs quantum well. A combination of PL, $I(V)$, and $C(V)$ measurements on these diodes, and on a similar control sample lacking the QD layer, indicates that the electronic states and charge distribution in this type of RTD are strongly affected by the presence of the InAs layer. When a bias is applied, marked changes in the QD PL intensity are observed. This effect is explained in terms of the capture into the dots of both electrons and photocreated holes, which are injected into the well by tunneling. The measurements show that this type of device can be used to induce controlled, resonant

\[ \text{FIG. 10. Comparison between the voltage dependence of the peak intensity of the dot PL, PL}_{\text{QD}} \]