Modulation of the luminescence spectra of InAs self-assembled quantum dots by resonant tunneling through a quantum well

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We investigate carrier dynamics in optically excited *n-i-n* GaAs/(AlGa)As resonant tunneling diodes that incorporate a single layer of InAs quantum dots in the center of the GaAs quantum well (QW). Voltage-tunable resonant changes in the dot luminescence are observed and are discussed in terms of the tunneling of carriers into the resonant states of the QW and of the capture of carriers from the QW into the dots.

Since the first experiments on resonant tunneling,[1] there has been great interest in the physics of this phenomenon and its potential application to high-speed electronics and optoelectronics.[2] The rapid advance in nanofabrication and growth of semiconductors has led to the development of new designs for resonant tunneling diodes (RTD's), including structures that incorporate self-assembled quantum dots (QD's). These are nanometer-sized clusters, which form spontaneously in strained semiconductor heterostructures.[3] In recent experiments, InAs QD's were either incorporated in a single AlAs tunnel barrier[4] or else in, or close to, a GaAs quantum well (QW) surrounded by two (AlGa)As barriers.[5,6] Tunneling spectroscopy has been used to study transport through the zero-dimensional electronic states of a dot[4,5] and to realize QD-based memory devices.[6]

In this paper, we investigate carrier dynamics in a series of optically-excited *n-i-n* GaAs/(AlGa)As double barrier RTD's in which a layer of InAs QD's is embedded in the center of the GaAs QW. We show that the photoluminescence (PL) emission of the dots depends strongly on bias, being affected by the capture into the dots of both electrons and photocreated holes, which enter the QW by tunneling processes. This effect, which relies upon the very short time for carrier capture into the dots compared to the dwell time of tunneling carriers into the well, provides a sensitive means of probing the tunneling dynamics of minority holes into the QW.

Our samples were grown by molecular-beam epitaxy. Structures $qd1$ and $qd2$ consist of two 8.3-nm $Al_{0.4}Ga_{0.6}As$ 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 was grown in the center of the well by depositing 1.8 (sample $qd1$) or 2.3 (sample $qd2$) monolayers (ML) of InAs. Undoped GaAs spacer layers of width 50 nm separate the $Al_{0.4}Ga_{0.6}As$ barriers from 2×10^{17}-cm^{-3} *n*-doped GaAs layers of width 50 nm. Finally, 3×10^{18}-cm^{-3} *n*-doped GaAs layers of width 0.3 μm were used to form contacts. For comparison purposes, a control sample (sample *c*) was grown with the same sequence of layers but with no InAs layer. The samples were processed into circular mesa structures of diameter 100 μm. A ring-shaped electrical contact was fabricated on the top of the mesa to permit optical access to the sample for PL spectroscopy and measurements of the current-voltage, $I(V)$, characteristics under illumination. The optical excitation was provided by the 633-nm line of a He-Ne laser.

Figure 1 shows the $I(V)$ characteristics of the devices in forward bias (negatively biased substrate) and without illumination. They differ substantially from each other: in the control sample (*c*) two resonances, $e1$ and $e2$, are observed corresponding to electrons tunneling through the first two quasibound states of the QW; in contrast, in samples $qd1$ and $qd2$, the low bias resonance is not observed and the second resonant peak is shifted to higher voltages. As a preliminary to interpreting these data, we first modeled the resonant states of this type of structure. We solved the Schrödinger equation in the effective-mass approximation for a GaAs well incorporating a two-dimensional InAs layer (i.e., no QD's) with thickness in the range 1–2 ML. The effect of the InAs layer is to lower the energy of the first QW resonant state ($e1$) below the GaAs conduction-band edge. In contrast, higher-energy resonant states are less strongly perturbed, particularly the odd-parity states, like $e2$, which have nodes near the center of the well.[7] This simple model neglects the real morphology of the InAs layer, which consists of dots and a two-dimensional InAs layer [wetting layer (WL)] beneath them. The effects of the QD morphology can be represented as local minima in the WL potential profile, producing a distribution of discrete and localized QD states below the continuum states of the WL. The effect of the InAs layer is to lower $e1$ below the GaAs band edge, thus making this state unavailable for resonant tunneling of electrons from the GaAs emitter layer. This is consistent with the observed suppression of the $e1$ resonance in $I(V)$ in samples $qd1$ and $qd2$.

The modification of the resonant states by the InAs layer also 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 the dots, at zero bias equilibrium is established by some electrons diffusing from the doped GaAs layers and filling the QD states. The result-

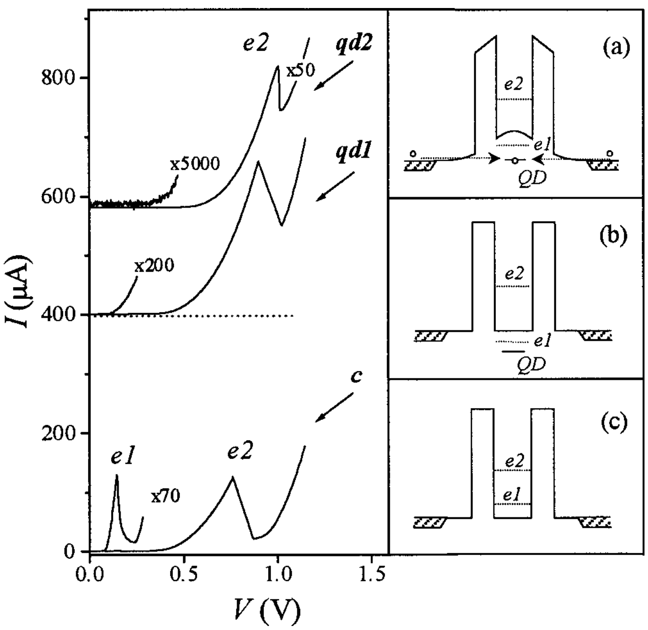

FIG. 1. $I(V)$ characteristics at 10 K for samples $qd1$ ($L = 1.8$ ML), $qd2$ ($L = 2.3$ ML), and c (no InAs layer). For clarity, the curves corresponding to the different samples are displaced along the vertical axis. $e1$ and $e2$ indicate the first and the second resonant current peaks in sample c. The insets show sketches of the RTD potential profile at zero bias for the control sample c and a RTD containing dots in the case of noncharged (b) and charged dots (a).

ing negative charge in the QW produces depletion layers in the region beyond the (AlGa)As barriers. The insets of Fig. 1 show schematically the RTD potential profile at zero bias for the control sample [inset (c)] and a RTD containing dots in the case of non charged [inset (b)], and charged dots [inset (a)]. We can estimate the areal density of electrons trapped in the dots from capacitance-voltage measurements:[8] at zero bias it is comparable to the QD density ($N_{\mathrm{QD}} \sim 10^{11}\,\mathrm{cm}^{-2}$) measured by atomic force microscopy on uncapped samples grown under similar conditions to samples $qd1$ and $qd2$. A significant voltage (~100 mV) is then required to reach the flat-band condition on the electron emitter side and to form the electron accumulation layer, from which electrons can tunnel into the QW. This shifts the voltage position of the $e2$ current peak in samples $qd1$ and $qd2$ with respect to sample c (see Fig. 1).

We now consider the effect of optical excitation on the $I(V)$ characteristics and the associated bias-dependent PL of the devices. We show that devices incorporating dots exhibit PL spectra, which differ considerably from those reported in the extensive literature on conventional RTD's.[9–12]

Figure 2 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 photoelectrons are swept by the depletion field into the positive contact (electron collector), whereas the holes move towards the electron emitter and form an accumulation layer adjacent to the right-hand barrier. During resonant transmission of electrons and holes through the well, some of the carriers can

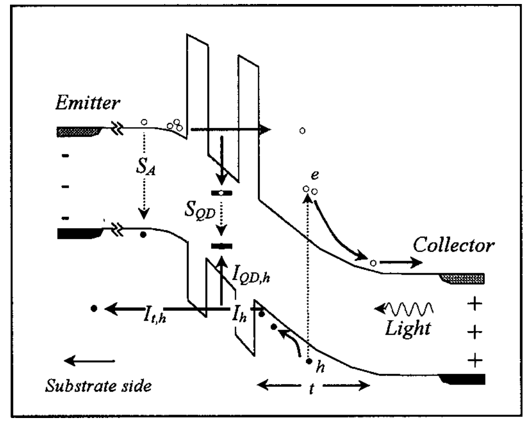

FIG. 2. Potential profile and carrier dynamics for a RTD in forward bias (negative biased substrate) with above-band-gap illumination. S_{QD} and S_{A} indicate the photoluminescence intensity for the electron-hole recombination in the dots and in the GaAs-doped layers, respectively.

be captured by the dots and then recombine radiatively. Carrier capture into QD's proceeds within times $\tau_c \sim 1$ ps,[13] much shorter that the characteristic dwell times (τ_d) of electrons and holes that tunnel resonantly into the well, but comparable to, or even longer than the semiclassical transit time (τ_t) of carriers through the QW and barrier region.[14] Due to this fast carrier capture, the bias dependence of the dot luminescence is thus very sensitive to the resonant tunneling process through the QW resonant states.

As shown in Fig. 3 for sample $qd2$, we observe an overall increase of current under illumination. This arises from the additional contribution to the current of the photocreated holes. The photocurrent increases rapidly at low voltages (<0.2 V), showing a resonant feature at ~0.15 V, it saturates as a function of V between 0.2 and 0.4 V, and then follows

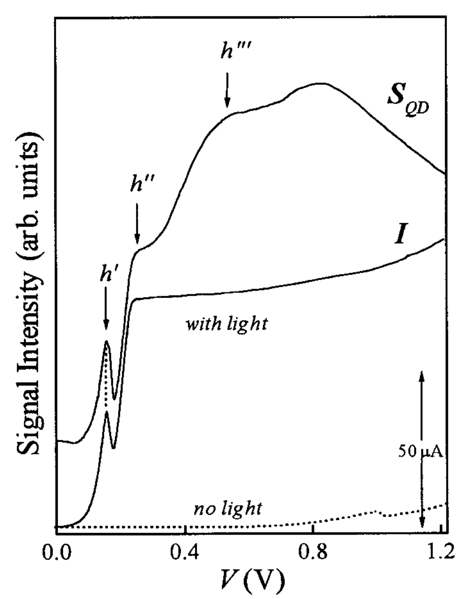

FIG. 3. Comparison between the voltage dependence of the current I with and without illumination, and of the dot PL intensity S_{QD} for sample $qd2$. The arrows indicate resonances (h',h'',h''') in S_{QD}. The diode is excited with above-band-gap laser light (633 nm) at a power density of $P = 20\,\mathrm{W/cm}^2$.

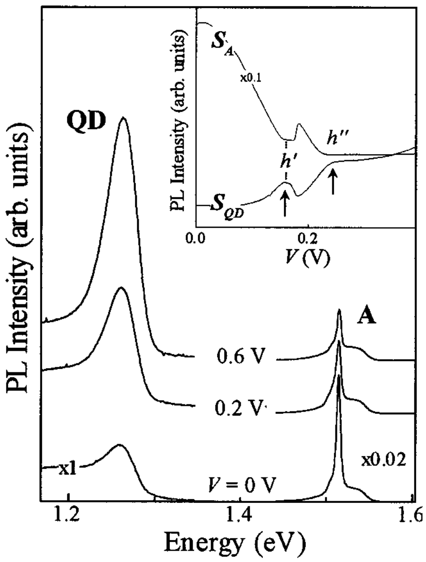

FIG. 4. PL spectra of sample $qd2$ at 10 K under different voltages. The bands QD and A originate from recombination of carriers in the dots and near-band-edge recombination in the GaAs-doped layers, respectively. The emission present on the low-energy side of the QD band arises from carrier recombination in the GaAs-doped layers and does not depend on bias. The inset shows a comparison between the bias dependence of the PL peak intensities, S_{QD} and S_A, of the QD and A bands. The diode is excited with above-band-gap laser light (633 nm) at a power density of $P=20\,\mathrm{W/cm^2}$.

nearly the same voltage dependence as the current measured in dark conditions. Note that the resonance $e2$ observed in Fig. 1 continues to be present, but is less well defined due to the contribution of the photocreated holes to the total current.

The PL spectra for sample $qd2$ are shown in Fig. 4 for three different voltages. The PL spectra show bands QD and A, which originate from recombination of carriers in the dots and near-band-edge recombination in the GaAs-doped layers, respectively. We do not observe any emission from the InAs wetting layer. However, this is not surprising. At low temperature, carriers preferentially recombine from the lowest-energy states available to them, namely, the QD levels.[15] The PL peak intensity of bands QD (S_{QD}) and A (S_A) exhibits resonant changes with bias. In particular, S_{QD} are S_A are in antiphase with each other: S_{QD} shows peaks, indicated by arrows, at the voltage where minima appear in S_A (see the inset of Fig. 4). The bias dependence of S_{QD} is compared with the $I(V)$ curve measured with and without illumination in Fig. 3. The $S_{\mathrm{QD}}(V)$ curve exhibits three clear resonant features (h', h'', h''') as indicated by arrows. One of these features (h') is also observed as a peak in the $I(V)$ under illumination.

The behavior of I, S_{QD}, and S_A can be explained as follows. The photocurrent depends on the carrier generation rate in the depletion region.[9] It increases with the incident optical power and with the thickness of the depletion region t, 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. The initial increase of photocurrent with bias is due to the increasing extent of the depletion region, where the screening charge is mainly due to the positive residual donors ($\sim 10^{15}\,\mathrm{cm^{-3}}$) in the nominally undoped GaAs layers. This increase saturates when the depletion region extends into the heavily doped ($2\times 10^{17}\,\mathrm{cm^{-3}}$) GaAs regions ($t\sim 50\,\mathrm{nm}$). Here the depletion width increases very slowly with bias, thus resulting in the saturation of current for $V \gtrsim 0.2\,\mathrm{V}$. 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 tunnel quickly into the well. This reduces the number of holes adjacent to the right-hand barrier. The resulting decrease in hole screening increases the depletion layer width 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 $I(V)$.

According to the carrier dynamics depicted in Fig. 2, we can understand the behavior of $S_{\mathrm{QD}}(V)$ and $S_A(V)$ qualitatively by considering the two components of the hole current, $I_h = I_{\mathrm{QD},h} + I_{t,h}$. Here $I_{\mathrm{QD},h}$ is the current associated with those holes that tunnel into the QW resonant states and are captured into the dots; $I_{t,h}$ corresponds to holes that pass through both tunnel barriers and recombine in the electron GaAs emitter layer. The relative strengths of $I_{\mathrm{QD},h}$ and $I_{t,h}$ are determined by the relevant tunneling time and the capture time into the dots. Using the device parameters, it is possible to estimate these times only approximately. Nevertheless the estimate provide a qualitative insight into the origin of the resonant structure in $S_{\mathrm{QD}}(V)$. When a hole tunnels through the two barriers nonresonantly, the time it spends in the QW is short and is given approximately by the time for a single transit of the hole across the well, $\tau_t \sim w/v_t \sim 10^{-13}\,\mathrm{s}$, where v_t is the hole transit velocity and w is the well width. In contrast, when a hole tunnels resonantly into a bound state of the QW, its dwell time τ_d is much longer and given by $\tau_t/T \sim 10^{-6}-10^{-8}\,\mathrm{s}$, where $T \sim 10^{-7}-10^{-5}$ is the transmission coefficient of the tunnel barriers estimated from the device parameters.[14] 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. 13) corresponds to a hole cross section for single dot of $\sigma_{\mathrm{QD}} \sim 10^{-12}\,\mathrm{cm^2}$, so that the probability of hole capture cross section on a single transit time is small, $\sigma_{\mathrm{QD}} N_{\mathrm{QD}} < 0.1$. Equivalently, the capture of a hole into the dot proceeds in a time $\tau_c \sim 10^{-12}\,\mathrm{s}$,[13] which is shorter than τ_d and longer than τ_t. These estimates provide a qualitative understanding of the origin of the resonant structure in $S_{\mathrm{QD}}(V)$. Off resonance, most of the holes make a single pass of the quantum well without interacting with the QD's, 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 large 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 $S_{\mathrm{QD}}(V)$ (see resonances h', h'', and h''' in Fig. 3). This description is confirmed by the observation of the weak antiresonant structure in S_A (see the inset of Fig. 4). This signal shows minima at the h' and h'' resonances, corresponding to a reduced number of holes recombining in the GaAs emitter layer due to the efficient hole capture into the dots.

The bias-dependent tunability of hole tunneling provides a means of varying the carrier density in QD's. This in turn affects the linewidth and energy peak position of the QD PL band. The energy peak position of the QD band follows the bias dependence of S_{QD} showing a maximum change of 6 meV. This may indicate a build-up of electrons and holes in the dots: as the number of carriers captured by the dots increases, the lower-energy QD states are saturated and higher-energy states start to be populated. Consistent with this state-filling effect, the QD band broadens slightly.

In conclusion, we have measured the electrical and optical properties of RTD's incorporating a single layer of InAs QD's in the QW. When a bias is applied, pronounced changes in the QD PL intensity are observed. This effect is explained in terms of the tunneling of minority carriers into the QW resonant states and subsequent carrier capture into the dots. The bias-controlled changes of the QD luminescence are consistent with a fast carrier capture into the dots and can be used as a means of probing the tunneling and capture dynamics of minority carriers.

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