Optically induced charge storage and current generation in InAs quantum dots

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(Received 31 October 2000; revised manuscript received 15 August 2001; published 25 January 2002)

We report on optically induced charge storage effects and current generation in self-assembled InAs quantum dots embedded in an InP matrix. Illumination with photons of energy higher than about 0.86 eV efficiently loads the dots with a maximum of about 1 hole/dot. The spectral response at lower photon energy is strongly enhanced at elevated temperatures. We present a detailed balance model for the dots and discuss the thermally assisted optical excitation processes pertinent to hole accumulation. We also show that these processes make the dots act as nanometer-scaled temperature-dependent current generators.

DOI: 10.1103/PhysRevB.65.073304 PACS number(s): 78.67.Hc, 78.55.Cr, 78.66.Fd

A self-assembled semiconductor quantum dot (SAD) is a nanometer-sized semiconductor box embedded in a matrix of another semiconductor material. The dots are naturally defined by the so-called Stranski-Krastanow[1] growth mode without any lithography steps. The formed dots can subsequently be capped with epitaxial quality barrier material in one single growth run without intermediate exposure of the interfaces. The dots' small size in conjunction with the difference in materials confine electrons and holes in all three dimensions in atomiclike states. During the last few years a lot of attention has been devoted to the growth and characterization of SAD's. The strong interest is motivated by their potential for future photonics and high-speed electronics, as well as by their intriguing electronic properties. To our best knowledge, the electronic structure of SAD's has hitherto been investigated primarily by photoluminescence spectroscopy and photoluminescence excitation spectroscopy.[2–4] More recently SAD's have also been addressed with absorption spectroscopy,[5,6] tunneling capacitance spectroscopy,[7,8] and space-charge spectroscopy, e.g., deep-level transient spectroscopy (DLTS) and photocapacitance.[9–13] The latter family of experimental techniques was traditionally invoked in the study of deep levels in semiconductors.[14]

So far, most studies have focused on InAs SAD's in GaAs and InP SAD's in GaInP. Considerably less attention has been given to InAs SAD's embedded in a matrix of InP. These dots are very interesting from the point of view of fundamental physics due to unusually large confinement energy for holes of about 0.43 eV (Ref. 11) and a complex electronic structure.[6] The dots are also suitable as active media in optoelectronic components compatible with the wavelength range of fibers (1.5–1.8 μm), e.g., temperature-insensitive lasers with low threshold current.

Another important application area for SAD's is in memory devices. Lundstrom *et al.* recently reported on an optical quantum dot memory capable of storing excitons for typically seconds at 3 K.[15] The large confinement energy of 0.43 eV for holes discussed in the present work facilitates memory devices where holes can be stored for hours even at 77 K.[11,16] In order to realize optical memory devices it is of course important to understand the relevant processes for optical charging of the dots. The present paper addresses this issue. The competing processes that actually govern optical charging, and the related current generation, in InAs dots buried in InP are discussed at length. Previous studies (some cited above) mainly address thermal emission rates of either electrons or holes confined to the dots, or the dots' electronic structure. As opposed, hereto we model the optical charging and current generation by simultaneously taking into account competing thermal and optical escape processes involving both the conduction and valence bands, as well as exciton recombination, and show that the model is in good agreement with experimental results for InAs dots embedded in InP.

The samples used in this study were grown by metal organic chemical vapor deposition. Details on the growth procedure can be found in Ref. 17. The dots are incorporated in the middle of a 4-μm not-intentionally doped (n type $\sim 10^{15}\,\mathrm{cm}^{-3}$) InP epitaxial layer. From atomic force microscopy (AFM) measurements of uncapped samples grown under similar conditions, we estimate a dot density of about $2 \times 10^{10}\,\mathrm{cm}^{-2}$. The dots are expected to be coherent, i.e., dislocation-free. Noncoherent dots under our conditions occur very infrequently and only show up as clusters for higher amounts of InAs deposition as shown in Fig. 1 of Ref. 17. Therefore, if no additional dislocations are inserted into the material during overgrowth, we can exclude dislocation effects. From AFM and optical measurements as well as from TEM studies of InAs dots we have no general indications that additional dislocations are created during overgrowth.[18] Also, using the experimental techniques employed in this work it is possible to get information on the spatial location of defects. We have no indications of specific defects situated at the site of the dots.

Another important question is the extent of group-V intermixing in the dots during epitaxial overgrowth. The intermixing phenomenon is complex and, to our best knowledge, no detailed analytical values for the here-investigated dots are available. However, there is a clear trend in the direction of highly As-rich material. InAs dots can easily be produced by a P-As exchange reaction when AsH_3 reacts with an InP surface. The As incorporation and replacement of P atoms is indeed dominating. A study of the effects of simultaneous introduction of AsH_3 (arsine) and PH_3 (phosphine) into the reactor was reported in Ref. 17 and it was found that the optical properties of the formed dots were almost identical as

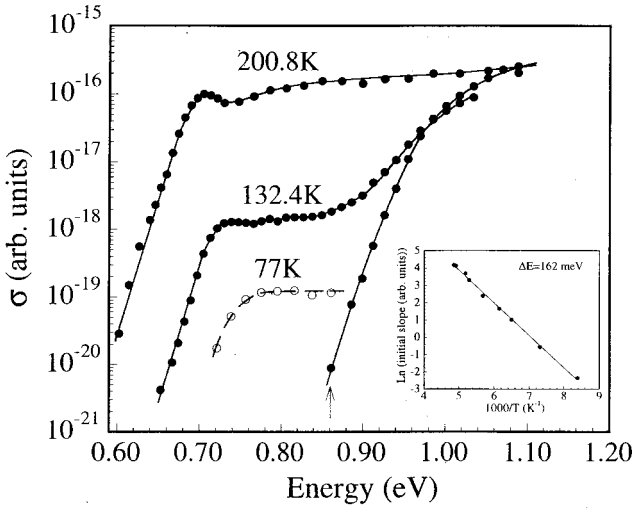

FIG. 1. Spectral distribution of σ measured at three different temperatures. The inset shows an Arrhenius plot of σ taken at the photon energy 0.775 eV. The arrow indicates the onset of electronic transitions from the dots' hole ground-state to the conduction band of the barrier.

when dots were grown with AsH_3 only. It should be mentioned that the high As/P incorporation ratio in general is known already since the fundamental investigations of Samuelson *et al.*[19]

Schottky junctions were prepared by evaporating a semitransparent 100-Å thick gold layer through a contact mask with circular apertures having a diameter of about 550 μm. The back ohmic contact was formed by alloying Au-Ge to the n^+-InP substrate.

In this work the optical properties of InAs dots were studied employing the photocapacitance technique. Optically induced charging of dots located in the space-charge region W of a Schottky junction result in changes in W that can be monitored by measuring the differential capacitance (more details can be found in Ref. 14). In general, illumination of our samples induces an initial steplike increase in capacitance, ΔC, followed by a slow transient. We show that ΔC corresponds to loading of holes in the dots, whereas the slow transient stems from current generated in the dots. We use traps situated in the space-charge region of the Schottky junction as internal probes to monitor these currents.

In Fig. 1 we have plotted the spectral distribution of the initial slope σ of the slow capacitance transient [i.e., $\sigma = dC(t)/dt|_{t=0}$], normalized with respect to the photon flux. Several important features implying that the photoresponse is related to the dots are readily noticed. A temperature-dependent threshold energy of about 0.60–0.65 eV is observed. The roughly exponential energy dependence of the photoresponse in the threshold region reflects the inhomogeneous broadening of the quantum dots. The peak observed at 0.70 eV in the 200.8-K spectrum is in excellent agreement with the fundamental transition observed in absorption.[6] At higher energy, a plateau region exists where σ is constant over an energy range of about 150 meV. The magnitude of σ in this region varies strongly with temperature. As an inset in Fig. 1 we show the Arrhenius plot of σ taken at the photon energy 0.775 eV. Interestingly, a perfect exponential dependence over three orders of magnitude is observed with an activation energy of about 0.16 eV. This activation energy is close to the ground-state energy of about 0.17 eV for electrons in InAs dots (measured relative to the band edge of the InP barrier) as reported by us in Ref. 11. Furthermore, there is a correlation between the optical response and the bias applied to the sample showing that the excitation process occurs at the spatial location of the dots. The observed photocapacitance signals are always positive. Since the material is weakly *n*-type the induced charge therefore has to be positive. It is tempting to attribute this charge to holes stored in the dots. We note however that neither illumination with low-energy photons ($h\upsilon > 0.43$ eV) nor increasing the temperature leads to the expected reemission of trapped holes back to the valence band of the InP barrier. Instead we found that the only way to remove the charge was to basically short-circuit the sample. This implies that the charge must be located much closer to the sample surface than the dot layer.

A model that can account for all the experimental facts discussed above is presented in the following. Resonant illumination with photons of energy smaller than the second threshold at about 0.86 eV (marked by an arrow in Fig. 1 and discussed later in the text) creates electron-hole pairs in the dots. The electrons and holes quickly thermalize to the ground state where after they escape to their respective band, or recombine depending on the temperature. The observed flat plateau region reflects the fact that the electrons and holes initially thermalize. The relevant electronic processes can principally be described by the rate equations

$$\frac{d\langle f_n\rangle}{dt} = G - R - \frac{\langle f_n\rangle}{\tau_n}, \tag{1}$$

$$\frac{d\langle f_p\rangle}{dt} = G - R - \frac{\langle f_p\rangle}{\tau_p}, \tag{2}$$

where G is the excitation rate, R the recombination rate, and τ_n and τ_p the time constants for emission of electrons and holes to the respective band. Both these emission time constants comprise an optical part and a thermal part, i.e., $\tau_{n,p} = [(1/\tau^o_{n,p}) + (1/\tau^t_{n,p})]^{-1}$. The optical emission time constant $\tau^o_{n,p}$ is obviously due to the excitation light (G) itself. Since the electrons are weakly bound, τ^o_n is also strongly affected by blackbody radiation from the surroundings. The importance of this radiation has previously been noted in defect spectroscopy on shallow impurities. The thermal emission time constants are governed by well-known Arrhenius expressions of $1/\tau^t_{n,p} = A\exp(-\Delta E_{n,p}/kT)$, where $\Delta E_{n,p}$ is the binding energy for the electrons and holes, respectively. The preexponential factor A is very large, primarily due to a large capture cross section, which makes thermal emission possible to observe in spite of the fact that $\Delta E_{n,p} \gg kT$. $\langle f_n\rangle$ and $\langle f_p\rangle$ are the average occupancies of the electron and hole ground states in the dots.

Under the present experimental conditions of weak excitation (μW) and $\tau_p \gg \tau_n$, $R \approx \langle f_n\rangle/\tau_r$, where τ_r is the exciton lifetime. The steady-state solutions to Eqs. (1) and (2) are

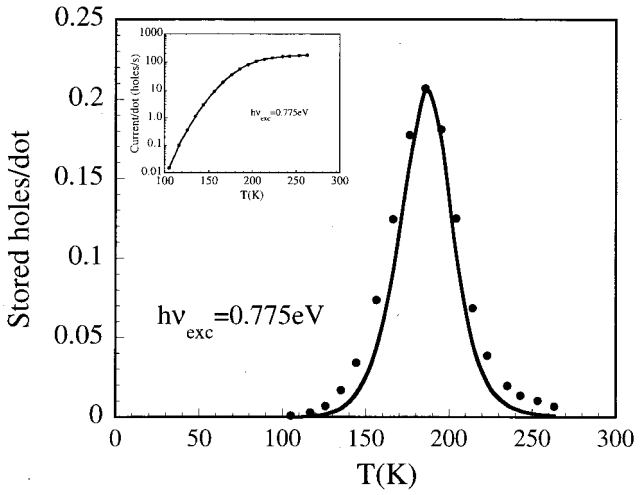

FIG. 2. Comparison between experimentally observed net accumulation of holes in the dots (solid circles) and those theoretically calculated (solid line). The inset shows the temperature dependence of the photocurrent generated in a single InAs dot.

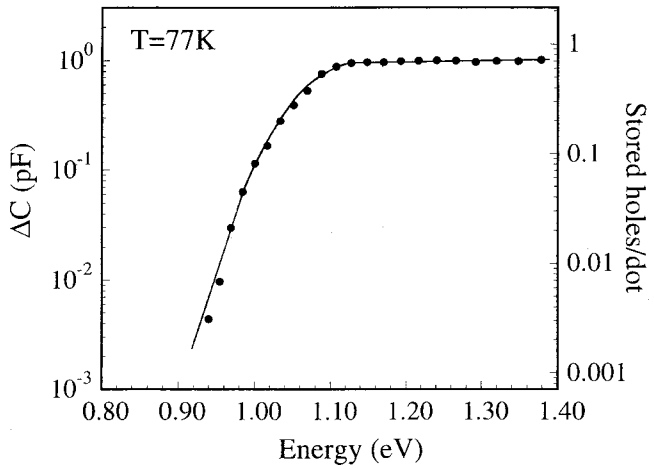

FIG. 3. Spectral distribution of the photocapacitance signal (left *y* axis) due to charging of the InAs dots and the corresponding average number of holes/dot (right *y* axis).

therefore given by $\langle f_n\rangle = G[\tau_r\tau_n/(\tau_r+\tau_n)]$ and $\langle f_p\rangle = G[\tau_r\tau_p/(\tau_r+\tau_n)]$. Effectively, each dot acts as a tiny temperature-dependent source of electron and hole currents, both with magnitude $I_{n,p}=qG[\tau_r/(\tau_n+\tau_r)]$. As an example, we plot the photocurrent due to a single InAs quantum dot excited with 0.775 eV as an inset in Fig. 2. For this plot G and τ_r were taken from simulations described below, whereas τ_n was experimentally determined. On their way to the surface of the sample the holes subsequently get captured by traps. This charging process is observed as a capacitance transient. We do not dwell on the possible origin of the traps except noting that DLTS measurements show that deep traps are present in the samples. For the plateau region observed at elevated temperatures in Fig. 1 is $\tau_r \ll \tau_n \approx \tau_n^t$ leading to currents, and thus capacitance transients, with a temperature dependence governed by ΔE_n in agreement with the inset. In contrast, at low temperatures the emission time constant for electrons is dominated by optical excitation, i.e., $\tau_n \approx \tau_n^o$. Since this source of excitation is relatively weak we would expect the corresponding current to be orders of magnitude smaller than actually observed (open circles in Fig. 1). We therefore attribute the low-temperature response to midgap background defects present in all our samples.

Besides acting as a minute current generator, each dot also stores a net amount of holes given by $\langle f_p\rangle - \langle f_n\rangle \approx \langle f_p\rangle = [\tau_r\tau_p/(\tau_r+\tau_n)]G$. The total accumulation of holes in all dots is experimentally detected as a small initial steplike increase in capacitance, ΔC. We verify that the signal indeed stems from accumulation of holes in the dots by noting that the capacitance signal vanishes when the sample is illuminated with low-energy photons as described previously (or if the temperature is raised). ΔC can be converted into the total amount of holes stored in all dots n, using the expression $\Delta C/C = nL/(W^2 N_D)$. Here C is the capacitance measured at the detection bias (-3 V in our case), L is the distance of the dot layer from the surface, and W is the width of the space-charge region of the Schottky junction. Since the average dot density is approximately known, $\langle f_p\rangle$ can readily be calculated. In Fig. 2 we plot the accumulation of holes for the typical excitation energy 0.775 eV (solid circles), together with the results from a fitting to the expression for $\langle f_p\rangle$ above (solid line). For the fitting procedure we have used emission time constants ($\tau_{n,p}$) extrapolated over many orders of magnitude from experimentally determined values.[11] Such an extrapolation is necessary since our setup cannot measure time constants smaller than about 1 ms. We therefore consider the agreement between theory and experiment to be good. It should be added that a perfect agreement could easily be obtained assuming that the effective energy barriers for electrons and holes become slightly smaller at high temperatures. This is not unreasonable if other processes such as, e.g., phonon-assisted tunneling become important. In the fitting procedure we obtained a generation rate $G=185$ electron-hole pairs/s and an exciton lifetime $\tau_r=2$ ns. We have also verified the expected dependence of $\langle f_p\rangle$ on the photon flux. At high temperatures where $\tau_p \approx \tau_p^t$ the accumulation scales with the flux Φ, since $G \propto \Phi$. At lower temperatures where $\tau_p \approx \tau_p^o$ the accumulation is basically insensitive to the flux since $\tau_p^o \propto 1/\Phi$. For all temperatures relevant to this discussion is $\tau_n \approx \tau_n^t$.

The second threshold at about 0.86 eV, observed at 77 K in Fig. 1 (indicated by an arrow), reflects an increase in photocurrent due to an electronic transition from the dots' hole ground state to the conduction band of the barrier. Adding the ground-state energy for holes of 0.43 eV and taking into account the influence of inhomogeneous broadening on the onset of the photoresponse discussed previously we obtain an energy in good agreement with the band gap of InP (1.40 eV). It is also possible that the excitation involves states in the wetting layer.[1] This layer, always present in the Stranski-Krastanow growth mode, is a thin strained quantum well with a thickness of only about 1 ML on top of which the dots are formed. In the barrier the electrons are efficiently swept away by the inherent electric field of the Schottky junction,

effectively leading to the case $\tau_r \gg \tau_n$ and a corresponding photocurrent $I_{n,p}=qG$. The photocurrent thus reveals the spectral response of G itself.

The increase in photocurrent in this energy range is accompanied by a large initial steplike increase in capacitance due to loading of holes in the dots. In Fig. 3 we plot the spectral distribution of this capacitance signal, converted into the average number of holes/dot. The onset is apparently slightly larger as compared to the 0.86 eV observed in Fig. 1. We attribute this discrepancy mainly to the fact that the signal in Fig. 3 has only been measured over two orders of magnitude. Interestingly, the spectral distribution becomes flat for photon energies exceeding about 1.10 eV. We understand this saturation in the following way: The absolute value of the capacitance signal gives the width of the space-charge region W from the relation $C=\varepsilon\varepsilon_0(A/W)$, where A is the area of the Schottky junction and $\varepsilon\varepsilon_0$ the dielectric constant. In reality, the edge of the space-charge region is not sharp due to the Debye tail, but still the approximation is expected to be good. In the experiment we observe that the saturation in accumulation occurs when W corresponds to the position of the dot layer. In other words, the charging of the dots results in a contraction of the space-charge region up to the point where electrons start to diffuse from the bulk into the dots, which render further loading of holes impossible. This model is furthermore supported by the fact that a slight decrease in the quiescent reverse bias (corresponding to a small change in W) instantaneously results in a decrease in the amount of holes stored in the dots.

In conclusion, we have reported on optically induced charge storage effects and current generation in self-assembled InAs quantum dots embedded in an InP matrix. Illumination with photons of energy higher than about 0.86 eV results in transitions from hole states of the dots to conduction-band states in the barrier that efficiently load the dots with a maximum of about 1 hole/dot. The spectral response at lower photon energy is strongly enhanced at elevated temperatures. We discuss this effect at length using a detailed balance model taking into account both thermal and optical emission processes.

The authors gratefully acknowledge financial support from the Swedish Natural Science Council, the Swedish National Board for Industrial and Technological Development, the Swedish Research Council for Engineering Sciences, and the Swedish Foundation for Strategic Research.

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