Carrier relaxation and electronic structure in InAs self-assembled quantum dots

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We studied the electronic structure and relaxation processes in InAs quantum dots embedded in GaAs. Using capacitance measurements along with photoluminescence spectroscopy, we estimate the energy splitting between the ground and first excited quantum-dot state in the conduction and valence band, respectively. There are five quantum-dot transitions observable in our photoluminescence (PL) spectra, which we attribute to allowed transitions between electron and hole states of the same quantum number. Phonon-related relaxation processes were studied combining PL, resonant PL (RPL), and photoluminescence excitation (PLE) experiments. In the RPL as well as in the PLE spectra, we observed enhanced signals at twice the phonon energies available in the system. Therefore, a maximum in the intensity of the PLE and RPL signal does not necessarily occur when most of the dots are pumped resonantly into an excited state. The main criterion, however, seems to be that the energy distance between the pumped levels and the levels below matches a multiple of the available phonon energies. Changing the pump power in our resonant PL experiments corroborates that at least in the small carrier density regime phonon-related processes are important for the carrier relaxation in InAs quantum dots embedded in GaAs bulk material. [S0163-1829(96)07039-7]

I. INTRODUCTION

The epitaxial growth of different materials on top of each other using molecular beam epitaxy MBE, metal organic chemical vapor deposition, etc. allows the creation of quantum structures with completely different electronic behavior compared to bulk material. Due to the thin deposited layers of different materials quantum effects are clearly observable. Devices with interesting properties have been developed by taking advantage of two-dimensional quantum phenomena.[1,2] Two-dimensional confined systems are relatively easy to fabricate with layer-by-layer epitaxy. It is much more difficult to obtain additional confinement in the lateral direction and achieve one-dimensional quantum wires[3] and zero-dimensional quantum dots (QD's).[4]

An easy way to fabricate zero-dimensional QDs is the growth of InAs on GaAs in the Stranski-Krastanow growth mode.[5] In this case a 1-ML-thick InAs layer is formed on GaAs at the initial stage of the growth. Due to the 7% lattice mismatch between GaAs and InAs, the two-dimensional growth changes into a three-dimensional one after the deposition of ≈1.5–1.6 ML InAs. Small InAs islands are formed. In this system zero-dimensional quantum phenomena have been clearly observed.[6–10]

The self-assembling QD structures are very interesting from both technological and physical points of view. A quantum-dot laser, for example, is predicted to have a very low threshold current because of its zero-dimensional density of states. On the other hand, for building a device it is very important to completely understand the physical properties of the QDs.

Much research has been done on these self-assembling QD's. However, the number and energetic positions of the energy levels[11,12] as well as the carrier relaxation processes in such QD's are still subjects of intense research.[13–22] So far the electronic structure is determined theoretically and confirmed by comparing the calculated transition energies with experimental photoluminescence (PL) data.[11,12,23] Size and shape effects on the dot potential, however, are not well known and understood. The situation becomes even more complicated because of the lattice mismatch between InAs and GaAs which produces a high strain in the system. Therefore, different theoretical assumptions lead to different models of the electronic structure in InAs self-assembled QD's. For example using a small pyramidal shape for the QD's the (PL) peaks under high excitation are explained by ''forbidden'' transitions between only one electron state and several hole levels.[11] On the other hand, a parabolic potential of a lens-shaped QD gives energetically equidistant transitions between electron and hole levels of the same quantum numbers.[23]

In our study we have determined the electronic structure of InAs QD's embedded in GaAs by combining capacitance (C-V) and photoluminescence spectroscopy. Therefore, no assumptions about the dot potential, the strain, and other parameters are necessary.

Having an idea about the electronic structure it is now possible to discuss the relaxation processes in self-assembled QD's. Several groups have already reported on this topic using either resonant PL (RPL) or photoluminescence excitation spectroscopy (PLE) to explore carrier relaxation processes in zero-dimensional systems.[13,14,21] The carrier relaxation seems to be dominated by phonon-related effects if the QD's are pumped resonantly. This has been confirmed by time-resolved measurements where the relaxation times in $Al_xIn_{1-x}As$ QD's were studied pumping on and off resonance with the phonon energy.[23] However, only relaxation into the ground state was studied and nothing was reported about the effect of different carrier relaxation efficiencies on the PL line shape pumping nonresonantly into the cladding layers. The carrier density should also have an effect on the relaxation mechanisms because at high carrier densities time-resolved experiments measured on $In_xGa_{1-x}As$ QD's suggest that other processes like Auger effects[16,19] might be responsible for the fast carrier relaxation in these dots.[15]

We have investigated carrier relaxation not only to the ground state but also between excited states comparing PL, RPL, and PLE spectra measured at the same sample position (i.e., the same dot size distribution and density). The line shape of the PL and RPL signals and the relaxation processes are discussed as a function of QD size and pump intensity. We were able to define and separate different relaxation mechanisms. In the PLE and RPL spectra various structures are observed which we attribute to enhanced luminescence caused by phonon-assisted relaxation. The difference between the energy of the pump light and the PLE and RPL structures, respectively, is in good agreement with the available phonon energies. Especially in the low carrier density regime it seems that phonon-assisted processes are an important carrier relaxation mechanism in InAs self-assembled quantum dots embedded in GaAs while at higher carrier densities other mechanisms like Auger effects become more and more dominant.

II. EXPERIMENT

We have grown two different samples in a Varian GEN II MBE system under an As pressure of 1×10^{-5} Torr. In both samples the substrate was not rotated during the InAs deposition. This procedure allows us to explore rapidly a range of dot sizes and densities grown simultaneously.

Sample I was especially designed for capacitance experiments. In this sample a 160-nm-thick GaAs/AlAs (2-nm/2-nm) short-period superlattice (SPS) was grown on a thick GaAs buffer layer at 600 °C followed by 20-nm Si-doped GaAs ($n_D=4\times10^{18}$ cm^{-3}). A 25-nm intrinsic GaAs layer separates the InAs dots from the doped GaAs layer. Before we started the deposition of InAs with a growth rate of 0.04 ML/s, we lowered the substrate temperature to 530 °C. The first 1.5-ML InAs were grown in 0.15-ML intervals followed by 2-s growth interruption. After that, we continued to deposit about 0.2-ML InAs in steps of 0.05-ML with 2-s delay time in between until the reflection high-energy electron-diffraction pattern changes from streaky [two-dimensional (2D) growth] to spotty (3D growth). After a short growth interruption we overgrew the InAs QD's with 5-nm intrinsic GaAs before the temperature was increased to 600 °C and an additional 25-nm GaAs was deposited. The whole structure was capped by a 116-nm-thick GaAs/AlAs (1 nm/3 nm) SPS and 4-nm GaAs to prevent oxidation of the AlAs layers. For our capacitance experiments we used conventional photolithography to fabricate circular gates (150-μm diameter). The Shottky interface is formed by a 15-nm-thick Cr layer, followed by 10-nm Ni and 200-nm gold for bonding the device. The back contact is a Ni-AuGe alloy annealed at 400 °C for 120 s.

Sample II is completely undoped, and was grown without growth interruption. The InAs dot region is embedded between two 1-μm-thick GaAs-AlAs (1.8 nm/1.5 nm) SPS's which were grown at 600 °C. The InAs QD region between the SPSs consists of ten periods of 1.8-ML InAs and 50-nm $In_{0.003}Ga_{0.997}As$ and was grown at a pyrometer reading of 530 °C. The 0.3% In concentration in the $In_xGa_{1-x}As$ spacer layer was used in order to exclude a flux transient during the InAs dot growth and ensure the same dot size and composition for each of the ten dot layers. Therefore, the ten InAs QD layers are as identical as possible. A 100-nm-thick $In_{0.003}Ga_{0.997}As$ layer separates the cladding layers from the dot region. These layers were grown at 600 °C. The whole structure is capped with 10-nm GaAs. Due to the 50-nm $In_{0.003}Ga_{0.997}As$ between the dot layers vertical alignment and electronic coupling of the QD's can be excluded.[24,25]

All C-V experiments were carried out at $T=4.2$ K using a 5210 EG&G dual phase lock-in amplifier. To measure the capacitance, an ac bias of 5 mV was added to a variable dc bias.[6]

For our PL studies we pumped our samples with an Ar^+ laser and a Ti-sapphire laser was used in our PLE and RPL experiments. Both lasers were focused down to a spot size of about 100 μm. The luminescence was dispersed by a 0.85-m double spectrometer, and detected by a cooled Ge detector with a spatial resolution of 0.5 nm.

A. Electronic structure of InAs self-assembled quantum dots

In order to obtain information about the various electron and hole levels in our QD's, we investigated sample I with C-V and PL spectroscopy. In this particular sample all dot levels are above the Fermi level only at an applied dc bias of $U\leqslant-1$ V. In this case the doping concentration in the n-doped GaAs region and the thickness of the intrinsic layers above determine the capacitance of the sample. With decreasing negative bias the electron ground state (labeled 0 in Fig. 1) is shifted below the Fermi level E_F. The QD's are loaded with one electron. This results in a change of the capacitance signal and a peak appears at $U=-0.85$ V in Fig. 1. The broadening of the peak is caused by a distribution in In-Ga composition, strain, and size of the QD's.

An additional energy is required to overcome the Coulomb blockade E_C if a second electron is added to the ground state of the dots. This process is described by the second peak at $U=-0.7$ V. The applied bias can be converted into energy according to[6]

$$E=\frac{t_0}{t_{\mathrm{tot}}}U, \tag{1}$$

where $t_0=25$ nm and $t_{\mathrm{tot}}=175$ nm are the thicknesses of the spacer layer and the total intrinsic region, respectively (see the inset of Fig. 1). From the distance between the two peaks we determined a Coulomb charging energy of $E_C=e^2/2C\approx20$ meV for InAs QD's embedded in GaAs. C is the self-capacitance of the quantum dot. The Coulomb charging energy of a metallic disk with radius 10 nm embedded in GaAs is $E_C=18$ meV which is in quite good agreement with our experimentally observed value. In order to put a third electron into the QD the first excited electron level (labeled 1 in Fig. 1) has to be shifted below the Fermi level, and the Coulomb blockade of two electrons has to be over-

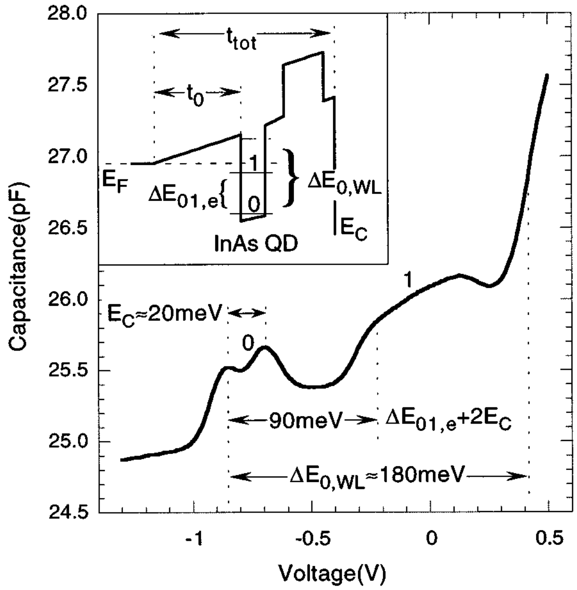

FIG. 1. Capacitance spectrum of sample I measured at $T=4.2$ K with an ac bias of 5 mV. The inset shows a schematic conduction band diagram (E_C) of the sample at $U=0$ V.

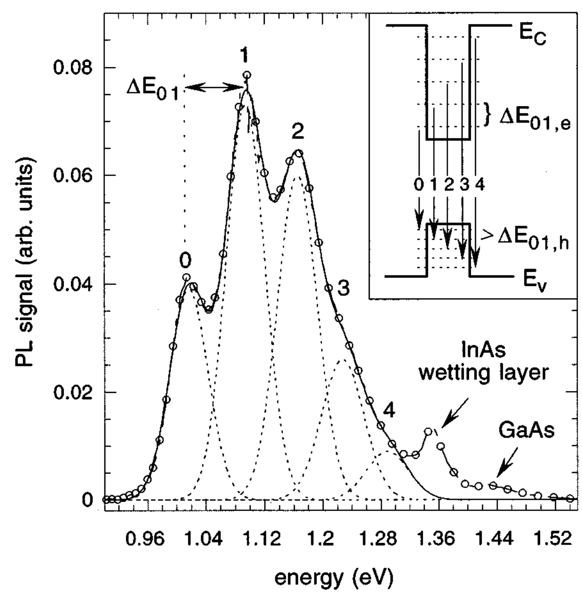

FIG. 2. PL spectrum (-○-○-) of sample I pumped with 30 W/mm^2 of an Ar^+-laser at $T=300$ K. The solid line shows a Gaussian line fit (---) to the experimental data. The numbered transitions in the inset are attached to the numbered PL peaks. E_C and E_V are the conduction- and valence-band edges, respectively.

come. This leads to an increase in the capacitance signal at $U=-0.25$ V. Taking the Coulomb effects into account we obtain an energy splitting $\Delta E_{01,e}$ between the ground and first excited states of roughly $\Delta E_{01,e}=90$ meV$-2E_C\approx50$ meV. Here it is assumed that E_C is independent of the number of electrons, which are already in the dot, and the QD level in which tunneling takes place. Despite our crude assumptions, $\Delta E_{01,e}$, determined with capacitance, is in good agreement with far-infrared data measured on the same or similar samples.[26,27] A more detailed study about Coulomb charging effects in terms of number of electrons in a quantum dot is given in Refs. 28 and 29.

The broad shoulder (labeled 1 in Fig. 1) at $U\approx0$ V describes the tunneling of the third, fourth, fifth, and sixth electron into the first excited electron state of the QD. This state consists of two degenerate energy levels ($m=+/-1$), and each can be filled with two electrons. The broadening is caused by Coulomb blockade effects. Finally the strong increase in the capacitance signal at about $U\approx0.4$ V corresponds to tunneling of the electrons into the two-dimensional InAs-wetting-layer states. With respect to Eq. (1) we determined a distance between the QD ground state and the wetting-layer ground state of roughly $\Delta E_{0,\mathrm{WL}}\approx180$ meV.

In order to obtain an idea about the hole confinement energies, we measured the PL spectrum of sample I pumped with 300 mW of a focused Ar^+ laser at $T=300$ K. In Fig. 2 the GaAs band edge as well as the InAs wetting-layer transition are observable at 1.424 and 1.35 eV, respectively. The QD PL signal at lower energies consists of five peaks which we attribute to transitions between electron and hole states with the same quantum numbers (inset of Fig. 2). According to Fig. 2 the PL peak labeled 0 reflects the recombination of an exciton in its QD ground state. The PL peak labeled 1 describes the recombination process of an electron in the first excited state with a hole in the first excited state (again these states consist of two degenerate energy levels with $m=+/-1$). The energetic difference ΔE_{01} between peaks 0 and 1 is equivalent to the sum of the energetic distance $\Delta E_{01,e/h}$ between the ground and the first excited states of the electron and the hole system. According to our model the peaks labeled 2–4 in Fig. 2 describe optical transitions between higher excited QD states. Due to Coulomb blockade effects, these higher excited states are not observable in the *C-V* spectrum of Fig. 1. Here the dots are loaded with electrons only, and the energy levels above the first excited state are shifted into the wetting-layer continuum.

However, in PL the total charge in the QD is 0, and therefore recombination processes between higher excited states are visible below the wetting-layer transition. This interpretation seems to be reasonable since both *C-V* measurements and far-infrared absorption show that at least four electron states can be associated with the QD's. This statement will be justified below. In addition, we assume that forbidden transitions do not contribute to the PL spectrum. As in capacitance spectra slightly different dots are again responsible for the broadening of the PL peaks. The energy splitting ΔE_{xy} of the peaks decreases from $\Delta E_{01}=77$ meV to $\Delta E_{34}=62$ meV with the increasing number of the transition, and their full width of half maximum is about 60 meV as deduced from a multiple gauss fit also shown in Fig. 2. As already mentioned above, the number of excited states is in good agreement with our results from capacitance spectroscopy. With the latter technique an energetic distance between the QD and the wetting-layer ground state of roughly $\Delta E_{0,\mathrm{WL}}\approx180$ meV is determined (see Fig. 1). The energy splitting between the ground and first excited electron states in the QD is about $\Delta E_{01,e}\approx50$ meV. Assuming the same energy splitting for all other electron levels, at least four electron states should exist in an unloaded InAs QD. However, the level splitting decreases with increasing quantum number, as can be observed in the PL spectrum of Fig. 2. Therefore, the existence of an additional electron level resulting in a fifth PL peak is quite reasonable.

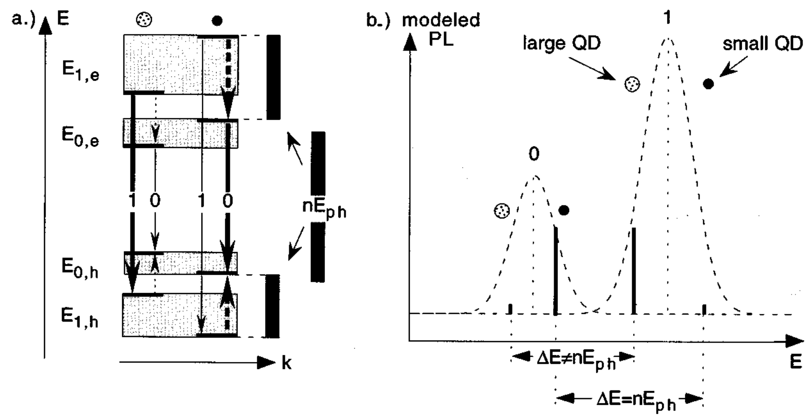

FIG. 3. Model of the phonon-related relaxation mechanisms (a) and of the resulting PL (b) in an ensemble of QD's with slightly different electronic structures. The gray boxes labeled $E_{0,e/h}$ and $E_{1,e/h}$ describe the energy-level distribution of the ground (0) and first excited (1) QD states in the conduction and valence bands, respectively. Dashed (full) arrows reflect the relaxation (radiative recombination) processes. The thickness of a line increases with the probability of a relaxation or a recombination process. The right (left) side of (a) depicts the situation for small (large) QD's marked with a small (large) disk. The black bar on the far right side represents the phonon energy available for the carrier relaxation, which splits into two parts used for electron and hole relaxation, respectively. The dashed Gaussian curves in (b) are the modeled PL of an ensemble of slightly different QD's without size-dependent phonon-enhanced relaxation taken into account. The full lines represent the PL signal of small and large QD's (marked with disks of different diameter) when size-dependent phonon relaxation is included according to (a).

The difference between $\Delta E_{01}=E_1-E_0=\Delta E_{01,e}+\Delta E_{01,h}$ (Fig. 2) and $\Delta E_{01,e}$ (Fig. 1 and the inset of Fig. 2) gives an energy splitting between the ground and first excited hole states of $\Delta E_{01,h}=27$ meV in our sample. In the considerations above, excitonic effects are less important because only energy differences are taken into account, and the binding energies for an exciton in the ground and first excited QD states are very similar.[30] According to Refs. 12 and 23, a ratio of 2 for the energy splitting of the electron and the hole levels seem quite reasonable.

B. Carrier relaxation in InAs quantum dots

Having an idea about the electronic structure in our QD's, it is possible to discuss the relaxation processes between different dot levels. Due to the energy dispersion in two- (wetting layer) and three-dimensional systems (GaAs cladding layers) hot carriers are able to relax down easily to the band edges using first optical and finally acoustic phonons.

Zero-dimensional QD systems have no energy dispersion but well-defined sharp levels in k space. If carriers want to relax down to a lower energy level (x) by phonon-related processes, the energy splitting $\Delta E_{xy}=E_y-E_x$ ($y=x+1$) between these levels has to match a multiple of the available phonon energies.[17] Since the electron and hole are localized at the same position in real space, the Coulomb interaction between these two types of carriers is very strong, and the electron and hole are no longer independent of each other. Therefore, excitonic relaxation might be the dominant thermalization process in QD's.[30,31] For fast phonon-enhanced carrier relaxation, either the energy splitting of the electron and the hole levels ($\Delta E_{xy,e/h}=E_{y,e/h}-E_{x,e/h}$) or a combination of both (excitonic relaxation) has to match a multiple of the phonon energies (E_{ph}):

$$\Delta E_{xy,e}+\Delta E_{xy,h}=nE_{\text{ph}} \quad (y=x+1,\ n \text{ integer}). \tag{2}$$

The phonon energies available in the system are 29.6 meV for the InAs WL and 31.9 meV for the InAs QD phonon, as well as 35 meV for the GaAs-InAs interface and 36.6 meV for the GaAs phonon.[21]

In a real experiment a huge number of QD's is investigated. All these QD's have slightly different ratios of the In-Ga concentration, strain, and size, which results in a distribution of the QD energy levels [represented by the gray boxes in Fig. 3(a)]. Larger QD's have their energy levels on the low-energy side [left side of Fig. 3(a) marked with a large disk], and smaller dots on the high-energy side of this distribution [right side of Fig. 3(a), marked with a small disk]. However, not only the absolute energetic position but also the energy splitting of the QD levels depends on the particular QD size [Fig. 3(a)].

Phonons act like size filters for these QD's. As already mentioned above, efficient carrier relaxation is possible only if Eq. (2) is fulfilled. Figure 3 shows that only the energy-level splitting $\Delta E_{01,e/h}=E_{1,e/h}-E_{0,e/h}$ in small QD's is equivalent to a multiple of the phonon energy nE_{ph} [black bar in Fig. 3(a)], while the splitting in large QD's is not. Therefore, carrier relaxation in large QD's [thin dashed line in Fig. 3(a)] is reduced compared to small QD's [thick dashed line in Fig. 3(a)]. Due to the size-dependent relaxation efficiency the intensity of the ground-state PL (0) of small QD's is higher compared to the signal of large QD's [Fig. 3(b)]. This results in an enhancement of the signal on the high-energy side of the modeled ground-state PL of Fig. 3(b). Since the confinement energy depends on the dot size in large dots, the level splitting does not match a multiple of the phonon energies. Therefore, the carriers stay longer in excited states and recombine there. Consequently the PL of excited states (1) is enhanced on the low-energy side for large dots (Fig. 3).

On the other hand, if the level splitting in large dots meets Eq. (2), the considerations above would result in an enhance-

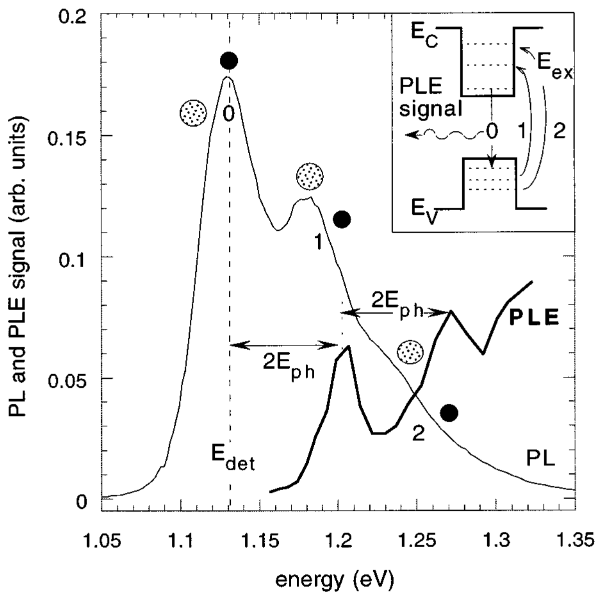

FIG. 4. PL (thin line) and PLE spectrum (thick line) of sample II measured at 10 K and pumped with an Ar^+ laser (50 W/mm^2) and a Ti-sapphire laser (27 W/mm^2), respectively. Analogous to Fig. 3(b), the energetic position of the PL signal is marked by large and small disks according to the QD size. The dashed line shows the detection energy E_{det} of the PLE signal. As soon as the energy of the pump laser (E_{ex}) is in resonance with the first (1) or second (2) excited-state transition (see the inset), and relaxation into the ground state (0) is possible, a signal appears at the detection energy.

ment of the ground-state PL on the low-energy side (large dots), and the PL of excited states is increased on the high-energy side (small dots). In any case, however, the PL does not necessarily reflect only the QD level distribution but is modified by size-dependent relaxation efficiencies if the splitting of the energy levels in the QD (i.e., the energetic distance $\Delta E = E_1 - E_0 = \Delta E_{01,e} + \Delta E_{01,h}$ between two QD transitions) is close to a multiple of the phonon energies. Such size-dependent relaxation effects should be also observable in the PLE signal of a QD ensemble. The PLE data do not only reflect the absorption of the sample, but in addition depend strongly on the carrier relaxation down to the detection energy.[21]

In order to study the relaxation processes in an ensemble of InAs QD's, we performed PL, PLE, and RPL experiments on sample II using a Ti-sapphire laser with 200-mW output power as a wavelength tunable pump source. Since we pumped resonantly with a single laser line, relaxation of a single exciton is studied with PLE and RPL experiments. According to Ref. 30 the energetic position of the energy levels (i.e., the transition energy) in a QD depends on the number of excitons in the dot. Consequently, as soon as an exciton is excited the QD levels change their energetic position. Thus, the transition energy comes out of resonance with the laser beam, and due to the zero-dimensional density of states in a QD excitation of a second exciton is suppressed. As soon as the first exciton recombines, the electronic structure of the QD recovers and it is possible to create the next exciton. A detailed theoretical study of excitons in self assembled QD is given in Ref. 30.

Figure 4 shows PL (thin line) and PLE (thick line) spectra of sample II measured at the same wafer position (i.e., the same dot size distribution and density). Due to the ten dot layers and the thick cladding layers the excitation per dot is significantly lower compared to sample I even at higher pump intensity. Therefore, in the PL spectrum of Fig. 4 the quantum-dot transitions at higher energies are less pronounced compared to the PL data of sample I in Fig. 2. Nevertheless, in the PL curve of Fig. 4 there are at least two excited QD states clearly observable at 1.18 and 1.25 eV, respectively.

The detection energy $E_{det} = 1.13$ eV of the PLE spectrum is marked by the dashed line. Due to the zero-dimensional density of states, the ground state of the selected QD's is not observable in PLE experiments,[8,9,21] unless a time-delayed experiment is used.[32] As expected from the PL data, the PLE signal shows two pronounced peaks which we attribute to transitions into the first and second excited QD states. As already mentioned above, a structure in the PLE spectrum is observed only if absorption of the laser light is possible, and if the excited electron-hole pair is able to relax down to the QD ground state.

In the PLE experiment shown in Fig. 4, the detection energy is equivalent to the energy, where the ground-state PL signal is maximum. Since the energetic distance between the PL transitions (0,1,2) is slightly smaller than twice the phonon energy E_{ph} mostly the small dots are investigated with PLE according to the size-dependent relaxation efficiency described in Fig. 3. In these QD's the carrier relaxation is most efficient, and their ground-state PL is at a maximum. Consequently a peak in the PLE signal appears on the high-energy side of an excited-state transition (Fig. 4). Only for such excitation energies E_{ex} are the carriers created in the proper dot size, and the carrier relaxation in these QD's is most efficient. Following the discussion in the previous paragraph, this results in a shift of the PLE with regard to the PL peaks (Figs. 3 and 4). The energy distance between the detection energy E_{det} (dashed line) and the first PLE peak as well as between the first and second PLE structures matches twice the phonon energies available in the system, which is a strong argument that phonon-related processes are very important for the carrier relaxation in InAs QD's.

To obtain more information about the phonons which are involved in these relaxation processes we performed resonant PL (RPL) experiments on the same sample position.[14] At an excitation energy E_{ex} of 1.157 eV [0 in Fig. 5(a)] electron and hole pairs are excited mainly into the ground state of small dots [the inset of Fig. 5(a)]. As already discussed those dots have slightly higher transition energies than larger dots and their ground-state PL is therefore on the high-energy shoulder of the PL peak at 1.13 eV. If there is no coupling between the dots, the carriers cannot relax down to lower energies, but have to recombine from the same state in which they had been excited. Therefore, the sample emits mostly at the excitation energy and the RPL signal at lower energies is weak in Fig. 5(a). With increasing energy the laser beam pumps more and more carriers into the first excited electron and hole level of the QD's. Thus relaxation into the ground state is possible which results in an increase of the RPL signal at lower energies. The relaxation process is most efficient in those QD's in which the electronic structure

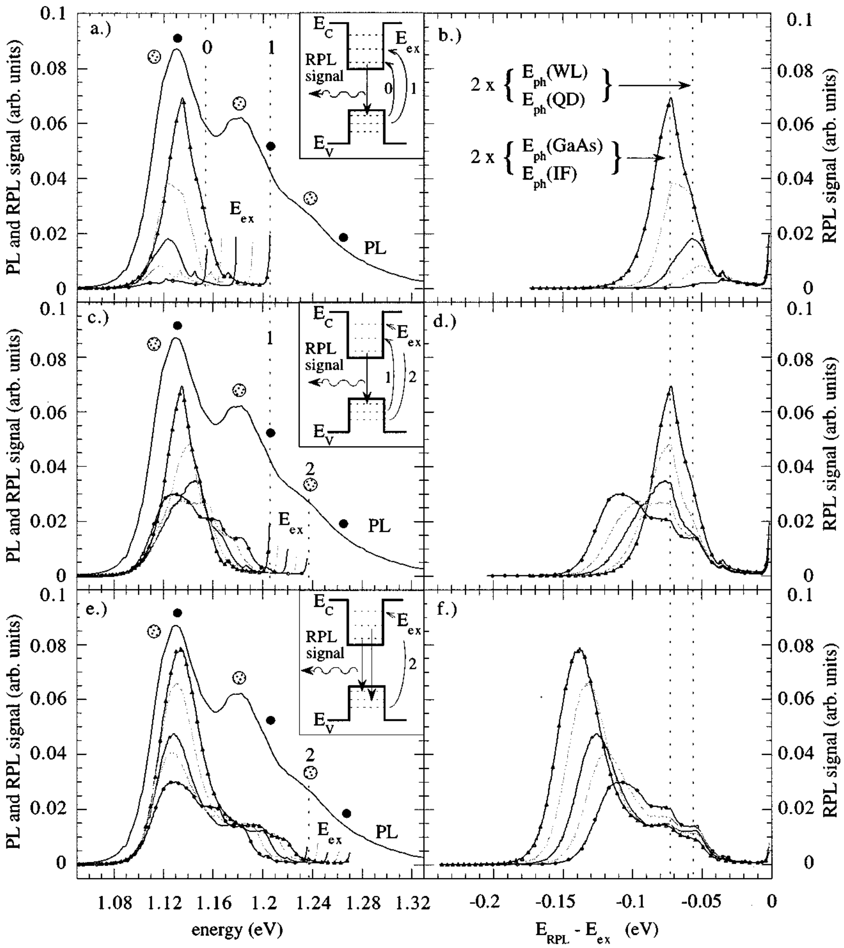

FIG. 5. PL and RPL spectra of sample II done under the same experimental conditions as in Fig. 4. The RPL spectra are measured at different excitation energies E_{ex} [(a), (c), and (e)]. The energetic position of the PL corresponding to the size of the QD is marked by large and small disks (compare Figs. 3 and 4). The inset shows the excitation and recombination for the labeled excitation energies. In (b), (d), and (f) the RPL curves are related to the excitation energy of the Ti-sapphire laser. The dashed lines mark the phonon energies E_{ph} which are available in the InAs-GaAs system.

fulfills Eq. (2). In this case the energy difference between the exciting and the emitted light matches a multiple of a phonon energy,

$$E_{RPL} - E_{ex} = \Delta E_{xy,e} + \Delta E_{xy,h} = nE_{ph} \quad (y = x+1,\ n \text{ integer}). \tag{3}$$

Following the previous discussion an enhanced RPL signal is observed in Fig. 5(a). At a pump energy of 1.207 eV [1 in Fig. 5(a) and Fig. 5(c)] most of the created carriers can relax down to the ground state via phonons and the intensity of the ground-state RPL signal reaches a maximum.

The phonon energies are independent of the excitation energy. Therefore, the energetic distance between a phonon-related RPL structure and the excitation energy has to be constant [see also Eq. (3)]. In fact, if the RPL signal is related to the excitation energy one of the peaks lines up at about twice the phonon energy of GaAs (36.6 meV) or the GaAs-InAs interface phonon (35 meV). A second one can be attributed to a relaxation process which is supported by the InAs QD phonon (30 meV) or the InAs wetting-layer phonon (29 meV) [Fig. 5(b)]. The broadening of the phonon structures is attributed to the high strain in the dot region which has also an influence on the phonon energies. We interpret the small peak at 35 meV as Raman-scattered laser light because its linewidth is only 3 meV, and its intensity shows almost no dependence on the energy of the pump light.

The intensity of the ground state RPL signal decreases for pump energies higher than $E_{ex} = 1.207$ eV and finally reaches a minimum at $E_{ex} = 1.237$ eV [2 in Fig. 5(c)] whereas a peak appears in the PL spectrum. In this case the carriers are pumped into the second excited state of larger dots. For most of these dots the splitting of the energy levels does not meet the multiphonon conditions for easy relaxation described in Eq. (2). As already mentioned above this causes an enhancement of the PL of the excited state and the ground-state RPL signal decreases. In addition to the ground-state luminescence a signal from the first excited state appears in Figs. 5(c) and 5(e). We attribute this effect to carriers which relax down from the second excited into the first excited state by multiple phonon-related processes [Figs. 5(d), 5(f), and the inset of Fig. 5(e)]. The splitting between the first excited and the ground state in these dots, however, does not match Eq. (2). Therefore, the probability of radiative recombination from the first excited state is enhanced. Due to the high strain and the strong QD confinement light-hole transitions are at much higher energies and can be excluded to be responsible for the PL signal at 1.18 eV in Fig. 5.[11]

With further increasing energy of the pump light more and more carriers are created in the second excited state of those dots, whose electronic structure fulfills Eq. (2) [Fig. 5(e)]. Since the relaxation efficiency is high in this case, the RPL signal of the ground state increases again. Due to broadening, phonon-related structures are no longer observable in

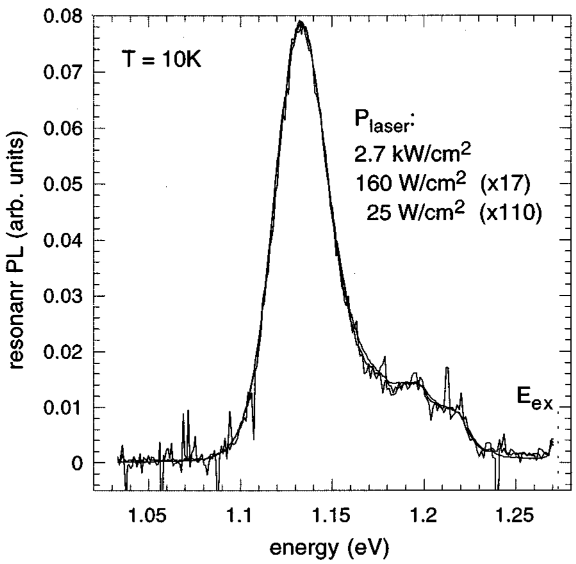

FIG. 6. RPL spectra of sample II measured at different pump intensities and with the same excitation energy E_{ex} of the Ti-sapphire laser. The spectra are normalized to the spectrum measured with the highest pump intensity (multiplication factors in brackets).

the ground-state RPL signal. The same phenomenon appears for the higher levels with further increasing pump energy. Finally the RPL signal becomes similar to the PL signal.

In order to rule out that saturation effects[13–15] of the lower levels are responsible for the RPL signal of the excited states we changed the pump intensity of the Ti-sapphire laser over two orders of magnitude. The integrated RPL signal changes linearly with the pump intensity but there is no change in the line shape observable (Fig. 6). This clearly indicates that our experiments were all done under equilibrium conditions and the characteristic line shape results from different relaxation probabilities between the energy levels in different QD's.

Following the previous discussion it is important to note that a maximum in the intensity of the PLE or RPL signal does not necessarily occur when most of the dots are pumped resonantly into excited states. The main criterion, however, is that the energy distance between the pumped levels and the levels below matches a multiple of the available phonon energies. This effect even appears in normal PL spectra. An efficient phonon-assisted carrier relaxation results in a carrier depopulation of the excited state, and the PL signal of this particular state is reduced compared to the signal of those dots whose energy-level splitting does not match the phonon energy. Such an effect causes a shift between the PL and PLE signals, as can be observed in Fig. 4.

Our results are confirmed by time-resolved experiments done on $Al_xIn_{1-x}As$ QD's.[20] In such structures a reduced relaxation time was observed if the $Al_xIn_{1-x}As$ QD's are pumped resonantly into an excited state which is separated from the ground state by one phonon energy.

III. CONCLUSION

We used capacitance and photoluminescence spectroscopy to study the energy splitting of electron and hole states in InAs self-assembled quantum dots embedded in GaAs bulk material. Taking Coulomb effects into account, we determined an energetic distance between the ground and first excited electron QD states of $\Delta E_{01,e} \approx 50$ meV. In combination with the PL data measured at the same sample this results in a level separation between ground and first excited hole QD states of roughly $\Delta E_{01,h} \approx 27$ meV. In our PL spectra measured with high excitation, we observe five peaks below the wetting-layer transition which we attribute to electron-hole recombination from quantum-dot levels of the same quantum number. The number of excited states is in good agreement with our results from capacitance spectroscopy. It seems that the Hamiltonian for an InAs QD system can be well approximated by a parabolic potential since the PL peaks are almost equidistant from each other. PLE and RPL experiments show clearly the existence of phonon-enhanced carrier relaxation into the ground and excited states if the energy splitting between the QD levels matches twice the available phonon energies. Saturation effects can be ruled out because a change of the pump power over two orders of magnitude has no influence on the line shape of the RPL spectra. Therefore, structures in the PLE and RPL signal do not necessarily depend only on the density of states but also on the efficiency of the carrier relaxation between the pumped level and the level below. In addition, different phonon-related carrier relaxation efficiencies for different QD sizes would also explain the slight shift between the PL and PLE structures observed in our experiments. Finally, it is important to keep in mind that the electronic structure as well as the relaxation processes are described in an easy model in order to give an idea about the situation in InAs self-assembled QD's grown at our growth conditions. However, much remains to be understood since different growth parameters (like temperature or As pressure) might result in the formation of different QD's with different energetic conditions, and the influence of carrier density on the relaxation processes is still unclear.

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