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Effects of Separate Carrier Generation on the Emission Properties of InAs/GaAs Quantum Dots

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ABSTRACT

Individual quantum dots have been studied by means of microphotoluminescence with dual-laser excitation. The additional infrared laser influences the dot charge configuration and increases the dot luminescence intensity. This is explained in terms of separate generation of excess electrons and holes into the dot from the two lasers. With increasing dot density and/or sample temperature, the increase of the luminescence intensity vanishes progressively, while the possibility to control the dot charge remains.

Semiconductor quantum dots (QDs), which effectively confine electrons (e's) and holes (h's) on the nanometer-length scale in all three directions, are considered widely as potential candidates for various optoelectronic applications such as QD lasers,[1] QD memory devices,[2] single-electron transistors,[3] and the generation of single photons with single quantum dots.[4] However, carriers confined in a QD are strongly interacting, which makes the energy states very sensitive to the exact number of the trapped carriers. Therefore, any operation of the QD-based devices is affected considerably by excess charge stored inside the QD,[2,3] which highlights the important role of the studies of multiparticle complexes with nonequal numbers of e's and h's in QDs.

Multiparticle complexes (excitons) consisting of an equal (neutral excitons) or nonequal (charged excitons) number of e's and h's have been studied extensively both theoretically[5−8] and experimentally.[9−18] In particular, the formation of negatively charged exciton complexes has been demonstrated in photoluminescence (PL) experiments.[6,9,10] The pioneer optical studies have been based on measurements of large ensembles of dots, so the results inevitably include the effect of inhomogeneous broadening, which prevents accurate measurements and hence a detailed understanding of the optical properties of the QD.

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However, in recent years the studies have been performed by means of micro-PL (μPL) spectroscopy on individual QDs. Such measurements are an extremely sensitive probe to the presence of excess charges in the QD, revealed by the fine spectral structure originating from different charged exciton complexes.[11,13,17,19−21] Several experimental approaches have been employed to load a single QD (SQD) with extra negative charges. Most commonly, contacted samples with an n-doped layer are employed. When an external voltage is applied, additional electrons will be transferred from the dopant layer to be captured into the dot.[15,16] The number of extra electrons in the QD can then be tuned by an applied external voltage. However, the applied voltage causes a shift of the entire spectrum and deforms the wave functions, which eventually leads to a change in the interaction energies. Also, more sophisticated sample designs have been used.[14] For instance, a layer of low-density In(Ga)As QD's was embedded in a wide GaAs quantum well (QW), while a neighboring more narrow QW was separated from the wide QW by a rather thin AlAs barrier, which serves as an effective supplier of extra carriers under optical excitation.

A third way to study charged excitons can be achieved by employing samples, where the QD's are initially filled with a random number of electrons from the background doping.[18] The drawback of this method is that the initial number of excess electrons depends on the unknown number of impurity atoms in the close vicinity of the investigated QD and hence is hardly controllable.

An alternative approach to creating and studying negatively charged exciton complexes in individual InAs/GaAs QDs was described in our earlier publication.[17] The existence of a well-defined threshold value ($h\nu_{th}$) of the excitation energy ($h\nu_{ex}$) was revealed in these experiments: if the excitation is shifted above this threshold ($h\nu_{ex} > h\nu_{th}$), then surplus e's will appear in the QD because of the ionization processes of shallow acceptor impurities located in the GaAs barriers in the vicinity of the dots. Consequently, this effect provides an effective tool for controlling the charge configuration of an individual QD by pure optical means.

A method complementary to controlling the charge state and the integrated emission intensity of the QD (I_{QD}) has been developed in our previous dual-laser-excited μPL experiments.[19] We have demonstrated that when the principal laser excitation is complemented with an infrared (IR) laser, the I_{QD} value increases several times (up to a factor of 5) compared to that of single-laser excitation. In addition, the QD charge state is altered in favor of a more neutral exciton configuration. However, in these initial experiments,[19] individual QDs were studied only at a fixed low temperature (T), 5 K.

In this paper we present further elucidation and verification of the novel mechanism proposed in ref 19 for increasing I_{QD} and altering the QD charge state by means of dual-laser excitation experiments. This is achieved by extended studies on the dependence of the investigated mechanism on the sample temperature and the dot density. It is shown that the effect of increasing I_{QD} progressively vanishes with increasing T as well as dot density. At the same time, it is observed that the possibility of altering the QD charge by changing the excitation energy remains for all of the temperatures and dot densities used.

The sample studied was grown by molecular beam epitaxy (MBE) on a GaAs (100) substrate. It consists of lens-shaped InAs QDs developed on an InAs wetting layer (WL) from about 1.7 monolayers of InAs deposited in the Stranski−Krastanov growth mode. The WL and the dot layer were sandwiched between two 100-nm-thick GaAs barriers. The sample was grown without rotation of the substrate, so a gradual variation of the In flux was achieved across the wafer, resulting in a gradient of the QDs density. The QDs were studied by means of conventional diffraction-limited μPL (a detailed description of the setup and the sample growth procedure is given in ref 11).

To excite the sample, we focused the beams from two Ti−Sp lasers on the same position of the sample surface down to a spot diameter of 2 μm. The excitation energy ($h\nu_{ex}$) of the main laser (L_0), could be tuned in the range from 1.410 to 1.530 eV with a maximum excitation power (P_0) of 20 μW. The IR laser, L_{IR}, operating at a fixed excitation energy, $h\nu_{IR} = 1.240$ eV, had its maximum output power (P_{IR}) of 100 μW. It is important to note that the energy, $h\nu_{IR}$, is well below the energy of any QD-related emissions and, accordingly, no signal from either the WL or the QDs was detected when exciting with only L_{IR}. The sample was positioned inside a continuous-flow cryostat operating at a temperature (T) from 5 up to 70 K. In this report, we present data measured on one specific SQD together with spots involving 2, 3, 6, or 50 QDs within the area of the laser spot.

A systematic μPL spectroscopy has revealed a very intriguing phenomenon, namely, the existence of a well-defined threshold energy ($h\nu_{th}$):[17] when exciting above or below this threshold (see arrows in Figure 1), completely different μPL spectra of the same SQD were monitored (as illustrated by the middle and right insets in Figure 1). Three predominant emission lines, marked as X, X-, and X- -, are detected. These lines have been interpreted earlier[11] in terms of excitons with different charge configurations, corresponding to the neutral, single, and double negatively charged excitons, that is, the 1e1h, 2e1h, and 3e1h charge states, respectively. Accordingly, when $h\nu_{ex}$ is increased above $h\nu_{th}$, the SQD charge configuration is effectively "switched" from the neutral to the negative state, as reflected in the μPL spectra by the complete quenching of the X line and instead a predominance of the X- - line (compare the middle and right insets in Figure 1, respectively).

The possibility to control the SQD charge state with a single laser, L_0, was ascribed to the effect of surplus e's generation from acceptors into the conduction band (CB) of the GaAs barriers[17] (arrow 3 in the left inset in Figure 1). Some part of the acceptors are expected to be filled with e's even at helium temperature as a result of e's capture from residual donors, which are inevitably present in MBE grown GaAs.[22] Consequently, at excitation with $h\nu_{ex} > h\nu_{th}$ surplus e's are excited (arrow 3 in the left inset in Figure 1), but electron−hole pairs (equal number of e's and h's) are also

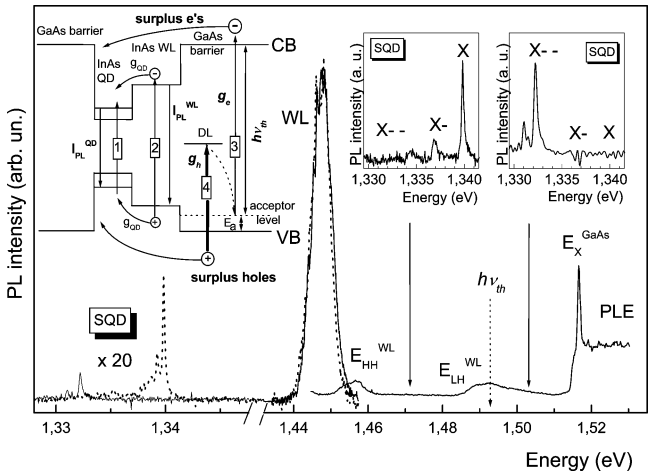

Figure 1. μPL spectra of a SQD and the WL measured at $T = 5$ K and $h\nu_{ex} = 1.503$ eV excited with single (solid lines) and dual (dotted lines) lasers, respectively, with an excitation of $P_0 = 40$ nW, $P_{IR} = 100$ μW, and $h\nu_{IR} = 1.240$ eV. The vertical dotted and solid arrows indicate the value of the threshold energy and excitation energy at which μPL spectra shown in the middle and right insets were recorded. The left inset shows the transitions involved in the energy scheme of the sample illustrating the positions of the conduction band (CB), the valence band (VB) of GaAs, and the acceptor level. The vertical and curved arrows are explained in the text. The middle (right) insets show the μPL spectra of a SQD measured at $T = 5$K, $P_0 = 40$ nW, and $h\nu_{ex} = 1.471$ (1.503) eV, respectively.

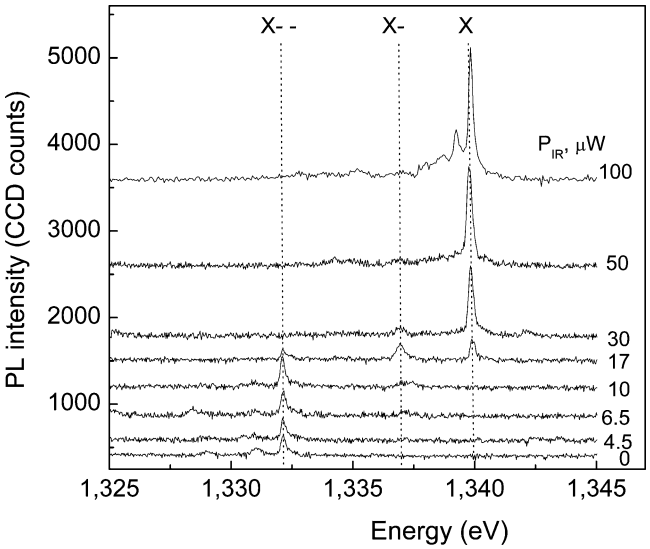

Figure 2. μPL spectra of the SQD, measured with dual-laser excitation with $P_0 = 20$ nW, $h\nu_{ex} = 1.503$ eV, $T = 5$ K, $h\nu_{IR} = 1.240$ eV, and for different powers on the infrared laser, P_{IR}, as indicated in the Figure.

excited as a result of the photons absorption in the WL (arrow 2 in the left inset in Figure 1). This explains the appearance of the predominantly "negative" charge configuration in the SQD at excitation with $h\nu_{ex} > h\nu_{th}$.

A spectacular effect was observed when, in addition to the excitation of L_0 with $h\nu_{ex} = 1.503$ eV $> h\nu_{th}$, the sample was illuminated with an infrared laser, L_{IR} (the μPL spectrum shown by the dotted line in Figure 1). Indeed, the μPL spectrum of the sample undergoes a dramatic change in this case: although the PL line of the WL remains almost unchanged, the SQD emission exhibits both a redistribution of the μPL spectrum in favor of the "neutral" exciton X line and an overall increase of its PL intensity, I_{QD}, by more than 5 times (dotted lines in Figure 1). The first effect is explained in terms of a generation of surplus h's, which are excited into the GaAs VB as a result of the IR excitation of e's from the GaAs VB to deep levels (DLs) in the GaAs band gap[23] (arrow 4 in the left inset in Figure 1).

The second effect with the huge increase of I_{QD} is similarly explained in terms of the separate generation of carriers by the two lasers: surplus e's by the L_0 and surplus h's by the L_{IR} laser (arrows 3 and 4 in the left inset in Figure 1). The fact that the carriers are generated separately has been justified by a number of experimental findings presented in ref 19.

In addition, the μPL spectra of the SQD, measured at a fixed laser power, P_0, exhibit a gradual modification with a progressively increasing P_{IR} as shown in Figure 2. Indeed, as P_{IR} (and consequently the number of extra holes) increases, the μPL spectra (Figure 2) exhibit a progressively increasing intensity, I_{QD}, accompanied by a smooth redistribution in favor of the X- and X spectral lines. These two experimental observations reflect a gradual compensation of the excess electrons accumulated in the SQD at single-laser-excitation conditions (i.e., $P_{IR} = 0$) by the extra holes generated by the IR laser.

Further, taking advantage of the gradient of the QD density, present in our sample, we have studied the behavior of the observed phenomena as a function of the number of QDs excited at a fixed laser energy, $h\nu_{ex} = 1.503$ eV $> h\nu_{th}$ at $T = 5$ K. This is achieved by changing the position of the laser excitation across the sample surface in such a way that there are 2, 3, 6, or 50 QDs within the area of the laser spot. It should be pointed out that the number 50 was estimated on the basis of the analysis of the absolute value of spectrally integrated PL signal, $I_{QD,}$ emitted from a QD ensemble at a given laser power, P_0. It was assumed that I_{QD} is directly proportional to the number of QDs located within the area of the laser spot, an idea that was verified from experiments on 1, 2, 3, and 6 QDs. It is found that the "multidot" μPL spectrum undergoes qualitatively the same changes under L_{IR} illumination as the SQD, that is, partly a "neutralization" and partly an increase of I_{QD}. However, the huge ($\sim$5.5 times) increase of I_{QD} recorded for a SQD is reduced progressively with increasing number of QDs to completely vanish for the highest dot density studied (50 QDs) (hereafter referred to as the multiquantum dots (MQDs)). The case of MQD excitation is represented in Figure 3, which shows μPL spectra recorded under single-laser (solid line) and dual-laser (dotted line) excitation.

To analyze this effect quantitatively we introduce a parameter β, which is the ratio of I_{QD} measured with dual-laser excitation (I_{QD}^{dual}) to I_{QD} recorded with single-laser L_0 excitation (I_{QD}^{single}), that is

$$\beta = \frac{I_{QD}^{dual}}{I_{QD}^{single}} = \frac{g_{QD} + g_{ad}}{g_{QD}} \quad (1)$$

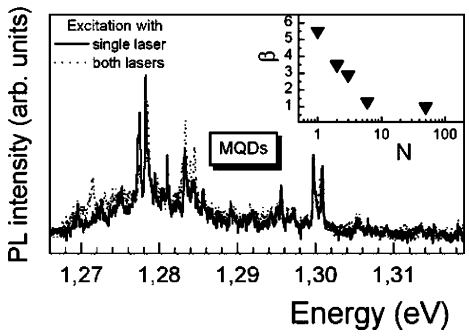

Figure 3. μPL spectra of MQDs measured at $T = 5$K with $h\nu_{ex} = 1.503$ eV. The spectra, excited by single (dual) laser(s), are shown by the solid (dotted) line for $P_0 = 40$ nW, $P_{IR} = 100$ μW, and $h\nu_{IR} = 1.240$ eV. The inset shows parameter β measured for a sample spot with different dot densities at $T = 5$ K, $h\nu_{ex} = 1.503$ eV, $h\nu_{IR} = 1.240$ eV, $P_0 = 40$ nW, and $P_{IR} = 100$ μW.

The experimentally observed fact that $\beta > 1$ means that additional generation of e−h pairs (g_{ad}) in the QD appears on top of the generation g_{QD} (shown by the solid curved arrows g_{QD} in the left-hand inset in Figure 1), achieved when exciting with only L_0. It is important to note that in order to increase the measured I_{QD}, both types of extra carriers (e's and h's) should be captured into the QD. Consequently, g_{ad} is determined by the smaller value of the generation rates of excess e's (g_e) and h's (g_h),[24] which, in turn, are determined entirely by the type and concentration of acceptors and DLs in the GaAs barriers but also by the excitation powers, P_0 and P_{IR}. Consequently, for given experimental conditions (referring to P_0, P_{IR}, and T) g_{ad} should have approximately the same value for the different sample spots where one, several, or many QDs were investigated. However, for increasing dot density, the PL signals collected from all of the QDs located within the area of the laser spot, that is, the value of g_{QD}, will increase. Therefore, taking eq 1 into account, one expects a progressive reduction of β with increasing number of dots (N) within the area of the laser spot. This prediction is in full agreement with the experimentally observed evolution of β with N shown in the inset of Figure 3.

To further assess the suggested model, we have studied the influence of temperature on the observed phenomena for excitation with a fixed photon energy, $h\nu_{ex} = 1.503$ eV > $h\nu_{th}$. Figure 4 shows the temperature evolution of the μPL spectra of a SQD measured with single (solid lines) and dual (dotted lines) laser excitation. For single-laser L_0 excitation, the μPL spectrum measured at $T = 5$ K is dominated entirely by the X- - line. In the T range between 5 and 25 K, a new line, redshifted with respect to X- -, appears. This line is tentatively identified as the triple-negatively charged exciton (the QD charge configuration 4e1h). The appearance of this exciton can be explained in terms of an increased carrier diffusivity[13] at elevated T, which leads to a more effective capture of e's into the QD with respect to the capture of h's.

However, at $T > 35$ K, the SQD becomes more neutral, as reflected in the redistribution of the μPL spectra in favor of the X- line. This evolution was tentatively investigated

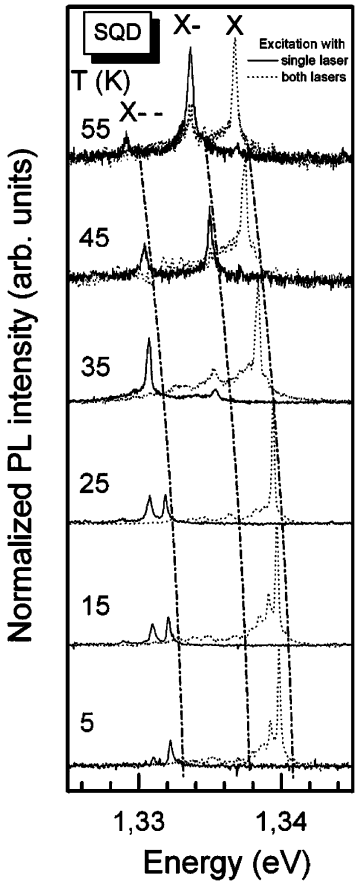

Figure 4. μPL spectra of a SQD, excited by single (dual) laser with $h\nu_{ex} = 1.503$ eV, $P_0 = 40$ nW, $P_{IR} = 100$ μW, and $h\nu_{IR} = 1.240$ eV are shown by solid (dotted) lines for a number of temperatures as indicated in the Figure. The dash−dotted lines are guides for the eye.

in our previous paper[17] and explained in terms of thermal ionization of acceptor holes to neutralize the excess electrons.

It should be stressed that there is a difference between the mechanisms of the compensation of the surplus e's by the thermally excited holes (taking place in the GaAs barriers) and by the holes generated by the L_{IR} (occurring inside the dot). This difference stems from the fact that thermally excited holes have a low kinetic energy (of the order of few meV's) with respect to the top of the VB of GaAs, whereas the optically created holes acquire a considerable kinetic energy of several hundreds of meV (equal to the difference between $h\nu_{IR}$ and the ionization energy of the DL[23]). Consequently, the latter holes are expected to be more mobile compared to the thermally excited holes. This fact eventually results in the accumulation of the optically created holes at the lowest energy level of the sample (i.e., in the QD), whereas the thermally created holes would rather be localized at the top of the VB of GaAs.

The addition of the IR laser results in a redistribution of the μPL spectra in favor of the X line (dotted lines in Figure 4) within the total range of T as well as in the progressive decrease of β with increasing T (Figure 4). The temperature dependence of β for a SQD is shown in Figure 5a for a number of P_0 values. Although β measured for $P_0 = 40$ and 80 nW reveals a similar behavior (a considerable decrease

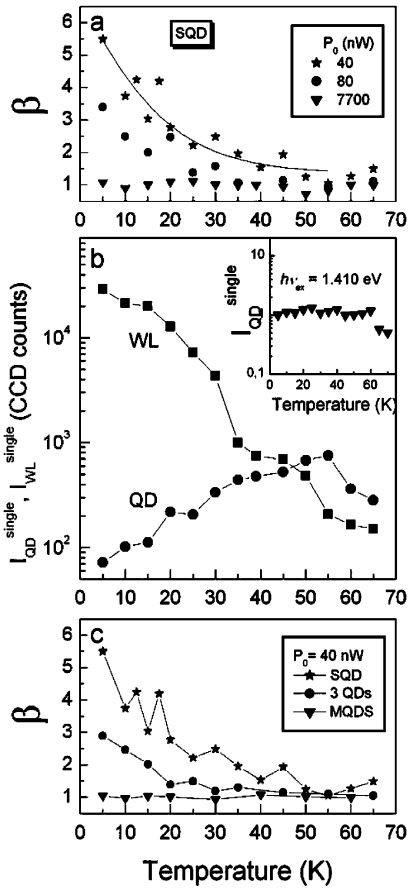

Figure 5. (a) Parameter β measured for a SQD at $h\nu_{ex} = 1.503$ eV, $P_{IR} = 100$ μW, $h\nu_{IR} = 1.240$ eV, for various temperatures and $P_0 = 40$, 80, and 7700 nW is shown by solid stars, circles, and triangles, respectively. The solid line corresponds to the result of calculations based on eq 1 as explained in the text. (b) I_{QD}^{single} measured for a SQD and I_{WL}^{single} recorded with a single-laser L_0 exciting at $h\nu_{ex} = 1.503$ eV, $P_0 = 40$ nW, and a number of temperatures shown by the solid circles and squares, respectively. The inset in b shows I_{QD}^{single} measured for a SQD excited with a single-laser L_0 at $h\nu_{ex} = 1.410$ eV, $P_0 = 17$ μW, and at different temperatures. (c) Parameter β measured at $h\nu_{ex} = 1.503$ eV, $P_0 = 40$ nW, $P_{IR} = 100$ μW, $h\nu_{IR} = 1.240$ eV, and a number of temperatures for a SQD, three QDs, and MQDs, shown by solid stars, circles, and triangles, respectively.

is observed already at $T = 35$ K), the case of high excitation ($P_0 = 7700$ nW) results in $\beta \approx 1$ for the entire temperature range studied. It should be pointed out that the above-mentioned compensation of the surplus e's with increasing T (which influences the value of g_e and, hence, g_{ad}) starts to play a role for $T > 35$ K, and thus cannot explain the considerable changes (reduction) of β observed in the temperature range $5 < T < 35$ K for high excitation.

To reveal the origin of the behavior of β, we measured the temperature evolution of the I_{QD}^{single} for the SQD (Figure 5b). Although the PL signal of the WL (I_{WL}^{single}) is reduced progressively (by more than 100 times within the total T range studied), I_{QD}^{single} exhibits an essential increase (by more than 10 times) when T is increased from 5 up to 55 K. A considerable decrease of the I_{WL}^{single} (Figure 5b) could be explained in the following way: at low T, the photoexcited carriers are captured into localized states of the WL and are located there until they recombine radiatively, to contribute to the I_{WL}^{single}. The existence of such localized states in the WL is evidenced by the observation of a number of sharp peaks (with the PL energy separation of <1 meV) superimposed on the low-energy tail of the WL emission band (Figure 1). As T increases, carriers become delocalized and their thermal velocity increases. Both of these effects result in a more efficient transport of carriers along the plane of the WL. Consequently, the probability for carriers to become captured by a QD or a center of nonradiative recombination (CNR) is increased considerably at elevated T. The increased thermal velocity of the carriers has two essential consequences: a quenching of I_{WL}^{single} because the CNRs will play a more important role, together with an enhancement of I_{QD}^{single} (as shown in Figure 5b). The crucial role of the carrier transport prior to the trapping into the CNR is described in detail in ref 25.

Our interpretation of the increased I_{QD}^{single} in terms of a temperature-facilitated transport of carriers is supported by the following experimental observation: for a L_0 laser excitation energy of $h\nu_{ex} = 1.410$ eV (i.e., excitation directly into the dot without any transport of carriers along the plane of the WL is needed prior to capture into the dot, shown by arrow 1 in the left inset in Figure 1), the PL signal of the SQD does not change in the temperature range $5 < T < 60$ K (see inset in Figure 5b). Consequently, the observed increase of I_{QD}^{single} (i.e., g_{QD}) with increasing T (Figure 5b) measured at a fixed value of g_{ad} (fixed values of P_0 and P_{IR}) should, according to eq 1, lead to a decreasing β. We have approximated the experimental points measured for I_{QD}^{single} in the temperature range of $5 < T < 55$ K in Figure 5b with a polynomial fit and substituted it into eq 1 (as shown by the solid line in Figure 5a). It fits nicely to the experimental points of β measured at $P_0 = 40$ nW. This fact strongly supports the proposed model. Accordingly, at high temperatures the only effect of L_{IR} (effect of the appearance of the surplus holes in the sample) on the μPL spectra would be the redistribution in favor of the X line. This prediction is in full agreement with the experimental observations shown in Figure 4. The experimental fact that $\beta \approx 1$ at $P_0 = 7700$ nW for the entire temperature range studied (Figure 5a) is obvious because g_{QD} already exceeds g_{ad} considerably at $T = 5$ K for such high values of P_0.

Figure 5c shows the T evolution of β measured at $P_0 = 40$ nW for a SQD, three QDs, and MQDs, respectively. The result for three QDs is similar to the dependence recorded for a SQD, whereas β for the MQDs remains ca. 1 within the entire T range. These experimental findings (Figure 5c) resemble the data measured for a SQD for a number of P_0 values (Figure 5a). Indeed, β for a SQD measured at $P_0 = 7700$ nW (Figure 5a) shows no T evolution, similar to β for the MQDs recorded at $P_0 = 40$ nW (Figure 5c). This is not surprising because in both cases g_{QD} already exceeds g_{ad} considerably at $T = 5$ K: in the first case it is due to the high value of P_0 used, whereas in the second case, it stems from the fact that the PL intensity detected for the MQDs is considerably higher than that for the case of a SQD.

Finally, we will rule out the alternative explanation of the observed effects induced by the IR laser as being due simply

to the sample heating as a result of the absorption of the IR irradiation with a high laser-power density (of ~2500 Wcm^{-2}) used in the present measurements. Indeed, under single-laser-excitation conditions, the increase of the sample temperature up to $T = 55$ K makes the SQD charge configuration more neutral (Figure 4) and, in addition, the total QD luminescence intensity, I_{QD}, increases by more than 10 times (Figure 5b). If the sample temperature was really increased in our experiments by the IR laser, then we would (according to Figure 5b) also detect an essential decrease (by more than 100 times) of the WL PL intensity, which is in contradiction to our observations shown in Figure 1. In fact, the WL emission band remains unchanged at both excitation conditions (single- and dual-laser excitation), which excludes any essential sample heating when the IR laser illuminates the sample.

In conclusion, an additional infrared laser employed in the dual-laser experiments with InAs/GaAs quantum dots supplies the sample under study with a certain amount of noncompensated holes. This fact considerably affects the μPL spectra registered from an individual dot: the charge state of the dot becomes more neutral and, in addition, the overall PL intensity can increase by as much as a factor of 5. The latter fact is explained in terms of an additive generation rate of excitons, g_{ad}, with respect to the generation provided by a single laser. The effect of increasing I_{QD} is demonstrated to vanish progressively with increasing sample temperature and/or the QD density. The effect of QD neutralization is, however, less influenced by these parameters. The revealed effects could be used in practice to manipulate the QDs charge state and emission intensity effectively.

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