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# Carrier transport in self-organized InAs/GaAs quantum-dot structures studied by single-dot spectroscopy

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A microphotoluminescence study of single InAs/GaAs quantum dots subjected to a lateral external electric field gives insight into carrier transport and capture processes into Stranski-Krastanov-grown quantum dots. The results obtained on the excitons in a single dot demonstrate a considerable luminescence intensity enhancement of the dot as well as a charge redistribution when an electric field is applied. The charge reconfiguration is evidenced by the transition from a predominantly negatively charged to a neutral charge state of the exciton. The model proposed to explain the charge redistribution is based on an effective hole localization at the potential fluctuations of the wetting layer.

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Semiconductor quantum dots (QDs) are often considered as potential candidates for optoelectronic applications such as QD memory devices,[1] QD lasers,[2] single-electron transistors,[3] and single-photon emitters.[4] The operation of these devices is based on an electric field involved across the dot structure, which highlights the importance of the effect of a field, external or/and internal, on the optical response of a QD.

Internal electric fields play an important role in many interesting optical phenomena such as two-color blinking,[5] spectral diffusion,[6,7] random telegraph noise,[8] and fluorescence intermittency.[9] For the effect of an external electric field, the investigations reported have primarily been treating a vertical field. The studies have essentially been focused on the capture and escape processes into and from the dot, i.e., the most essential factors for the optical characteristics of a dot.[10–12] However, such experiments do not allow studies on the step prior to capture, i.e., the carrier transport in the plane of the wetting layer (WL).[13]

In order to gain information on this lateral carrier transport, the carriers should be exposed to a lateral electric field. However, only a very limited number of such studies can be found in the literature: CdSe/ZnSe, GaAs/AlGaAs, and InAs/GaAs QDs have been exposed to a lateral electric field,[14–16] but the experimental conditions did not allow probing of the associated carrier transport in the presence of a lateral field. Indeed, in Ref. 15 the single QDs (SQDs) were excited by resonant pumping directly into the QD's excited states, while in Ref. 14 a special sample design (in the form of a mesa with a lateral size of 50 nm) containing an individual QD was used. As a result, a redshift of the QD's microphotoluminescence ($\mu$PL) lines and a decrease of the SQD PL intensity with an increasing external lateral electric field have been revealed.[14,15] From investigations of an ensemble of InAs QDs exposed to a lateral electric field, conclusions could be drawn only on the strong change in the oscillator strength of the transitions studied.[16]

In this paper, we report on a study of the effect of an external lateral field on the transport properties, as probed by the optical response of a single dot in microphotoluminescence. Experimental conditions are employed in which a laser beam was focused on the sample surface down to a spot diameter of 2 $\mu$m (still considerably larger than the lateral size of the SQD of 35 nm). The $\mu$PL results, obtained for various excitation powers ($P_0$) and temperatures ($T$), demonstrate a striking dependence of the PL intensity as well as the charge state on the external voltage ($V_{dc}$). According to the proposed model, the deficit of holes in the QD at zero field ($V_{dc}=0$ V) is caused by a more effective localization of holes at potential fluctuations of the WL during the lateral transport. At $V_{dc}>0$ V, the holes may tunnel out of the localizing potentials to become captured into the QD, effectively neutralizing its charge state. Our results demonstrate that the transport properties of photoexcited carriers should be taken into account for optical experiments with InAs/GaAs QDs to understand the origin of the emission lines from QDs measured at ordinary conditions, i.e., without any external fields.

The samples studied were grown by molecular beam epitaxy on a semi-insulating GaAs (100) substrate. The buffer layer was prepared with a short-period superlattice $40\times 2$ nm/2 nm AlAs/GaAs at a growth temperature of 630 °C. On top of a 100 nm GaAs layer, the QDs were formed from a 1.7 monolayer thick InAs layer deposited at 530 °C. A first growth interruption of 30 s. was used to improve the size distribution. Then the dots were covered with a thin GaAs cap layer with a thickness of $t_{cap}=3$ nm before a crucial second growth interruption of 30 s. Finally a 100 nm thick GaAs layer was deposited to protect the QDs. As a result, lens-shaped InAs QDs were developed with the height and diameter of 4.5 and 35 nm, respectively.

The dot density was extremely low with an average distance between the adjacent QD's of around 10 $\mu$m, which allowed excitation of a single QD in a diffraction-limited $\mu$PL setup. A lateral electric field is derived between two In gate electrodes with a 15 $\mu$m spacing on top of the sample. By excitation from a tunable Ti-sapphire laser, with an excitation energy ($h\nu_{ex}$) in the range from 1.23 to 1.77 eV, the luminescence signal was monitored by a nitrogen-cooled

 

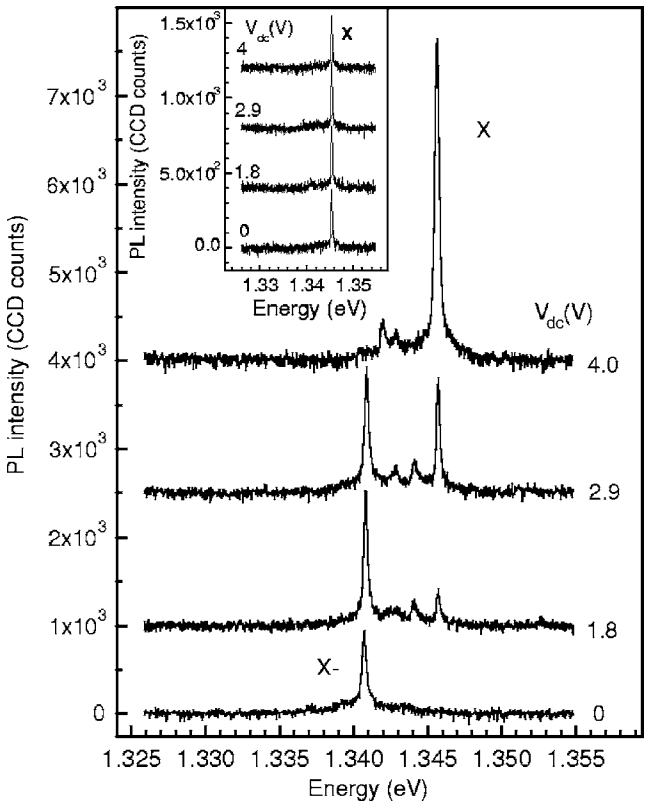


FIG. 1. $\mu$PL spectra of a single QD measured at $T=5$ K, $P_0=20$ nW, $h\nu_{ex}=1.675$ eV, for a number of biases, as indicated in the figure. The inset shows $\mu$PL spectra of the QD measured at similar conditions, but for an excitation energy of $h\nu_{ex}=1.40$ eV.

charge-coupled device CCD camera allowing a spectral resolution of 0.15 meV. Eight single dots at different spatial positions were examined with a resulting analogous behavior.

The spectrally integrated PL intensity of the QD, $I_{QD}$, is approximately 1% relative to the corresponding intensity of the WL, to be compared with the corresponding volume ratios of $\approx 10^{-3}$. Consequently, the PL signal from a QD is primarily not determined by the absorbing dot volume, but is rather due to transport and capture processes of carriers from the WL into the QD. This conclusion is further emphasized by the fact that the $\mu$PL spectra exhibit no emission from GaAs, even for above GaAs band gap excitation.

Also the charge state of the dot, i.e., the relative population of electrons ($e$) and holes ($h$), is essentially determined by these transport and capture processes, as elucidated by the electric-dependent measurements. The two excitonic $X^-$ and X lines (Fig. 1), predominant in the $\mu$PL spectra at low excitation powers, are identified as negatively charged and neutral exciton recombination emissions from the SQD, which correspond to the SQD charge configurations of ($2e1h$) and ($1e1h$), respectively. This interpretation is based on the following.

(i) Detailed theoretical calculations[17] show that for the case of a lens-shaped QD (as in our study), the spectral line corresponding to the SQD charge configuration of $2e1h$ ($1e2h$) should appear red- (blue) shifted with respect to the line of the neutral exciton ($1e1h$ charge configuration in the SQD). The reverse situation is predicted for the cone-shaped QD.[17] The progressive reduction of the transition energy of the emission from negatively charged exciton complexes in a lens-shaped QD with increasing number of extra electrons (up to 20) was also calculated.[18]

(ii) The characteristic trion (i.e., singly negatively charged exciton) redshift of ~5 meV in the InAs/GaAs QD system has been reported recently in the experiments of other authors (see Ref. 11 and references therein).

(iii) Photoexcited carriers, which migrate in the plane of the WL prior to capture into the SQD, will, with a certain probability, become localized at potential fluctuations of the WL, which are due to the growth-induced variations of alloy composition and strain along the plane of the WL.[19,20] The probability for carrier localization (in other words, its activation energy) is higher for holes due to their larger effective mass. Consequently, it is predicted that the lateral transport of holes will be hindered compared with the electrons. As a result, a more probable capture for electrons (compared with the capture of holes) into the SQD is expected, which should result in a "negative" charge configuration in the SQD (i.e., a predominant $X^-$ line in the $\mu$PL spectrum at $V_{dc}=0$ V). On the contrary, at excitation directly into the SQD ($h\nu_{ex}=1.40$ eV), i.e., in the case where no transport of photoexcited carriers occurs prior to capture into the SQD and hence, the capture probabilities of electrons and holes into the SQD are equal, the $\mu$PL spectrum of the SQD entirely consists of the $X$ line (inset in Fig. 1). It should be stressed that at excitation energies below the WL, crossed transitions (between the WL continuum and the QD-confined states) can play an important role.[21,22] In general, these transitions may result in different probabilities for electrons and holes to be captured into the SQD and, as a consequence, the $X$ line (inset in Fig. 1) might have been interpreted as a charged exciton complex instead of a neutral exciton. As was shown in Refs. 21 and 22 the influence of crossed transitions on the absorption of QDs is entirely determined by the excitation energy used. However, our data (not shown here) reveal that in a broad (50 meV) range of excitation energies below the WL, the $\mu$PL spectra obtained consist of the same $X$ line, which supports our interpretation of the $X$ line as being the neutral exciton.

(iv) To create the charge configuration of $2e1h$ in the SQD absorption of more than one photon (each of which creates only $1e$ and $1\ h$ in the sample) is needed. Consequently, a superlinear dependence of the amplitude of the $X^-$ PL line on the excitation power ($P_0$) is predicted. The inset in Fig. 2 a shows the spectrally integrated intensities of the $X^-$ ($I_{X^-}$) and $X$ ($I_X$) lines in their dependence on $P_0$. Both $I_{X^-}$ and $I_X$ could be approximated with $P_0^m$, where $m$ is found to be 1.16 and 1.00, respectively.

At unbiased conditions, the $X^-$ line dominates the $\mu$PL spectrum of the single QD (Fig. 1), but for an increasing bias, the $X$ line appears and successively gains intensity. In addition, a significant increase of the total dot PL intensity $I_{QD}$ by up to a factor of 4, is observed. As stated above, the photoexcited carriers will migrate in the WL prior to capture into a dot. However, this migration is sensitive to WL potential fluctuations caused by growth-induced variations in alloy

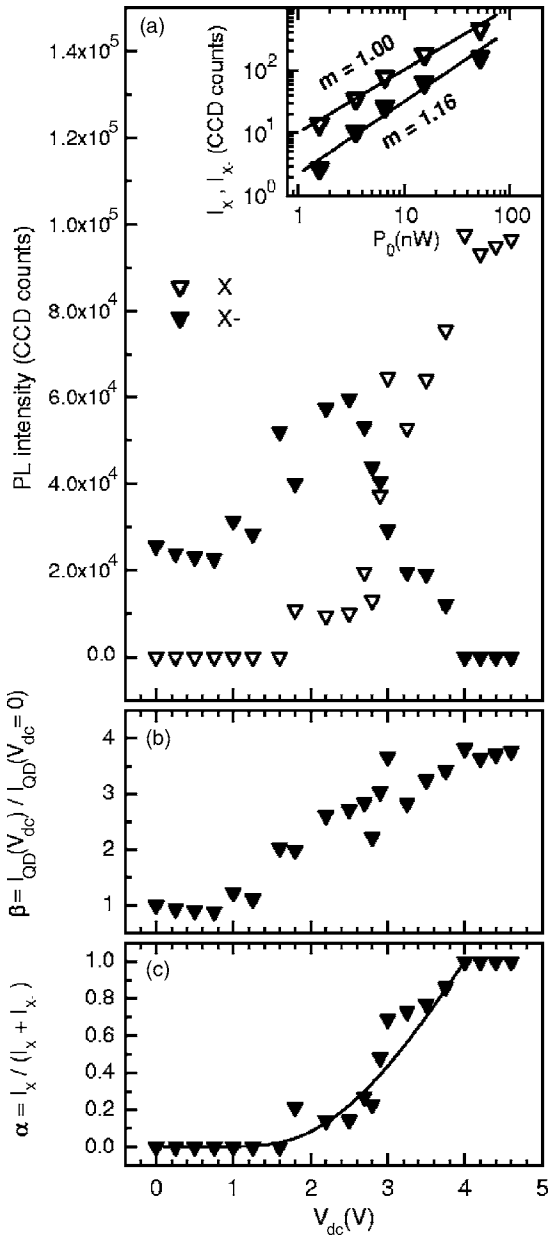


FIG. 2. The dependencies of (a) $I_X$ and $I_{X^-}$, (b) $\beta$, and (c) $\alpha$ on $V_{dc}$, measured at $T=5$ K, $P_0=20$ nW, and $h\nu_{ex}=1.675$ eV. The solid line in (c) is a fit to the model described in the text. The inset in (a) shows the dependence of $I_{X^-}$ and $I_X$ on $P_0$ measured at $V_{dc}=0$ and 6 V, respectively, at $T=5$ K. The solid lines in the inset represent fits to the data based on the method of least squares.

composition and strain, since capture of the carrier(s) will occur with a certain probability.[19,20] Consequently, the lateral transport is anticipated to be more efficient for electrons than for the heavier holes, which in turn will instigate a higher capture rate of electrons into the QD and a predominant $X^-$ exciton, in accordance with our experimental observations (Fig. 1).

The role of the internal electric field, created by charged impurities in the vicinity of the dots, has been elucidated earlier.[23] In the low-bias domain, this internal electric field will dominate over the applied electric field and just minor PL amplitude variations of the $X$ and $X^-$ lines ($I_X$ and $I_{X^-}$) are observed [see Fig. 2(a)]. However, for a higher bias ($0.7<V_{dc}<1.6$ V), an enhancement of $I_{X^-}$ is observed in the $\mu$PL spectrum (reaching a maximum at $V_{dc}=2.4$ V), which is explained in terms of an increasing drift velocity for the delocalized carriers. For an even higher bias $V_{dc}>1.6$ V, the neutral $X$ exciton appears with a progressively increasing PL intensity $I_X$ [Fig. 2(a)]. It should be stressed that the total integrated dot PL intensity ($I_{QD}=I_X+I_{X^-}$) increases monotonically by almost a factor of 4 in the range $0.7<V_{dc}<4$ V, as illustrated in Fig. 2(b) by the $\beta$ parameter, defined as $\beta=I_{QD}(V_{dc})/I_{QD}(V_{dc}=0)$. This observation is in striking contrast with the directly excited dot (with below-WL excitation when no carrier transport occurs prior to carrier capture into the QD), and $I_{QD}$ remains unchanged in the entire bias range studied (inset in Fig. 1).

The charge redistribution in favor of the neutral exciton $X$ observed in the $\mu$PL spectra for higher bias (Fig. 1) necessitates an increased probability for holes to get captured into the dot. An alternative explanation based on a tunneling process of excess electrons out of the dot, triggered by the bias, is excluded, since a decrease of $I_{QD}$ with increasing bias should be expected for excitation both above and below the WL energy, which is in contradiction with our experimental results [Fig. 2(b) and inset in Fig. 1]. Instead, the following model for the charge redistribution is proposed. The prevailing deficit of holes at zero bias (see, e.g., Fig. 1) is explained in terms of an enhanced probability for localization of holes relative to electrons in the WL potential fluctuations. These potential fluctuations will be considered as one-dimensional potentials along the direction of the external electric field. Under an applied electric field, holes can tunnel through the barrier of these potentials with a tunneling rate of $R_T$. The delocalized hole can, in a subsequent step, migrate and become captured by a dot, with a certain probability $\gamma_h$ to neutralize the charge state of the dot (with a capture rate of $R_h^{loc}=\gamma_h R_T$). Consequently, the initially negatively charged dot can be neutralized, if a delocalized hole is captured into the dot within a time interval that is shorter than the time between two successive events of electron capture into the dot, i.e., $R_h^{loc}>R_e^{opt}$, where $R_e^{opt}$ corresponds to the generation rate of photoexcited electrons into the dot.

In order to assess this neutralization process quantitatively, a parameter $\alpha$, defining the degree of neutrality, $\alpha=I_X/(I_X+I_{X^-})$, is introduced [see Fig. 2(c)]. According to the model proposed, an elevated temperature is expected to result in a more neutral charge configuration, even at $V_{dc}=0$ V, since the localized holes are assumed to become thermally released, in accordance with experimental observations of the $\mu$PL spectra (Fig. 3) or by the increasing parameter $\alpha$ (inset in Fig. 3).

It should be noticed that the bias-induced charge redistribution detected at $T=5$ K (Fig. 1) is also observed at any higher temperature, provided there is an initial negative charge configuration (i.e., an observable contribution from the $X^-$ exciton) at $V_{dc}=0$ V (Fig. 3). Furthermore, the observed bias-induced enhancement of the total intensity $I_{QD}$ is progressively reduced with increasing temperature. This fact is likely explained in terms of an increased delocalization of the carriers achieved at elevated temperatures also at zero

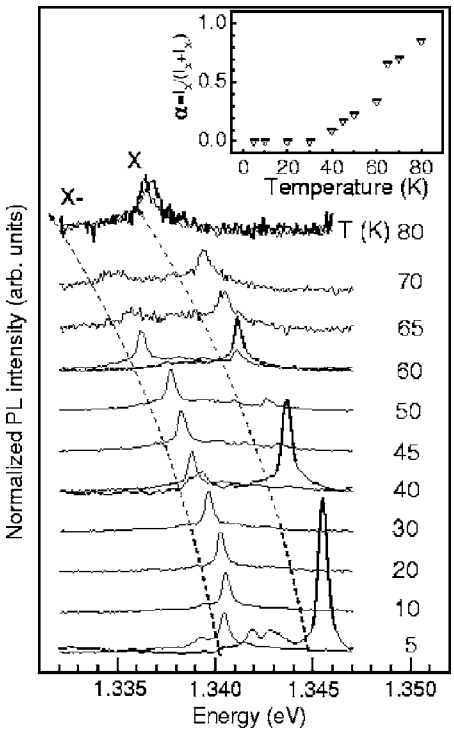


FIG. 3. The thin (thick) lines represent $\mu$PL spectra of the QD measured at $V_{dc}=0$ V (6 V) and $P_0=40$ nW, $h\nu_{ex}=1.675$ eV for a number of temperatures, as indicated in the figure. The dotted lines are guides for the eye. The inset shows $\alpha$ as a function of the temperature at $V_{dc}=0$ V as calculated from the spectra in the main figure at $V_{dc}=0$ V.

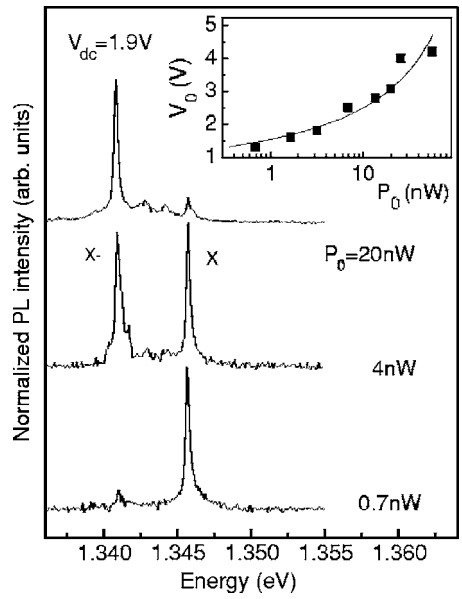


FIG. 4. $\mu$PL spectra of the QD measured at $h\nu_{ex}=1.675$ eV, $V_{dc}=1.9$ V, $T=5$ K for different excitation powers $P_0$, as indicated in the figure. The inset shows $V_0$ as a function of $P_0$ measured at $h\nu_{ex}=1.675$ eV and $T=5$ K. The solid line represents the result of calculations based on Eq. (1).

bias together with a considerably increased thermal velocity of the carriers, as reflected by an enhancement of $I_{QD}$ by a factor of 3 as the temperature is increased from 5 to 50 K without any electric field applied.

In the next step, we will pursue a quantitative comparison of the experimental results on the degree of neutrality $\alpha$ [as deduced from Fig. 2(c)] with the ratio $R_h^{loc}/R_e^{opt}$ according to the suggested model. It should be stressed that the dot intensity $I_{QD}$, which is described by the optical rate $R^{opt}$ at $V_{dc}=0$ V, is determined by the generation rate of minority carriers, i.e., holes. Consequently, $R^{opt}=g\gamma_h$ and $R_e^{opt}=2R^{opt}$ (because the dot is on time average occupied by two electrons and one hole), where $g=P_0 d_{GaAs}k_{abs}/h\nu_{ex}$ is the laser pumping rate, $d_{GaAs}=200$ nm is the thickness of the two GaAs barriers, and $k_{abs}=1.8\times10^4$ cm$^{-1}$ is the GaAs absorption coefficient at $h\nu_{ex}=1.675$ eV.[24] To calculate $\gamma_h$, we have assumed that (i) only $\approx$40% of the photoexcited carriers take part in the radiative recombination,[23] (ii) $I_{QD}$ is only $\approx$1% of the WL PL intensity (as stated above), and (iii) the dependence of $I_{QD}$ on $V_{dc}$ is given by $\beta(V_{dc})$ [Fig. 2(b)].

With these assumptions, the neutralization probability $\gamma_h(V_{dc})=4\times10^{-3}\times\beta(V_{dc})$ can be evaluated and a generation rate of $R^{opt}=4.1\times10^8$ s$^{-1}$ is obtained (for $P_0=20$ nW and $V_{dc}=4$ V). The validity of this estimate is confirmed by the appearance of the biexciton in the $\mu$PL spectra at $P_0>100$ nW (which corresponds to $R^{opt}>2\times10^9$ s$^{-1}$ according to these calculations). In fact, the biexciton should appear when the carrier generation rate into the dot exceeds the exciton recombination rate, which has been reported to be $(600\text{ ps})^{-1}=1.7\times10^9$ s$^{-1}$ for InAs/GaAs QDs.[25] One can then express $\alpha$ as

$$\alpha=\frac{I_X}{I_X+I_{X-}}=\frac{R_h^{loc}}{R_e^{opt}}=\frac{\gamma_h R_T}{2\gamma_h g}=\frac{1}{2g}\frac{h}{4m^*L^2}\exp\left(-\frac{8\pi\sqrt{2m^*}}{3ehF}E_i^{3/2}\right) \tag{1}$$

where $F$ is the electric field applied across the localization potential, $E_i$ is the ionization energy of a hole from this potential and $R_h^{loc}=\gamma_h R_T$, where the hole tunneling rate $R_T$ from a localization potential of width $L$ is derived from Ref. 10. A fitting procedure of the input data by Eq. (1) [the solid line in Fig. 2(c)] demonstrates that the experimental points agree satisfactorily with the dependence predicted by the suggested model.[26] From the slope of the solid line, an activation energy of $E_i=2.8$ meV can be deduced. Consequently, thermal release of holes out of the confining potential fluctuations at elevated temperatures ($T>30$ K) is expected and is consistent with the temperature dependence of $\alpha$ (inset in Fig. 3). In order to achieve a completely neutral configuration, i.e., $\alpha=1$ at $V_{dc}=4$ V, an extension of the localization potential of $L=47$ nm is required according to Eq. (1), which is in reasonable agreement with the reported lateral dimensions, which however vary significantly, from a few tens up to several hundreds of nanometers.[20]

The tunneling rate $R_T$ is, as expected, dependent on the bias $V_{dc}$, but is also strongly influenced by the excitation power $P_0$ [via $g$, as follows from Eq. (1)]. Accordingly, for a given bias, $R_h^{loc}>R_e^{opt}$ at low power $P_0$, i.e., a neutral charge configuration is expected, while for a higher power, $R_h^{loc}<R_e^{opt}$, which will result in a negatively charged dot. It is consequently anticipated that, for a fixed $V_{dc}$, an increasing

power $P_0$ will result in a charge redistribution from neutral to negative, as is experimentally confirmed by the transition from the $X$ to the $X^-$ exciton in the $\mu$PL spectra (Fig. 4). Based on the model proposed, the intensities of the $X$ and $X^-$ excitons can be balanced with equal amplitudes (i.e., $\alpha=1/2$) by a proper choice of voltage $V_0$. This fact is illustrated in the inset in Fig. 4, showing the power dependence for maintaining equal intensities of the $X$ and $X^-$ lines as a function of the applied voltage $V_0$. These experimental results are compared with the predicted power dependence as evaluated from Eq. (1) (with $\alpha=1/2$) and found to be in nice agreement (see inset in Fig. 4).

In conclusion, the importance of the carrier transport characteristics for the luminous efficiency of self-organized InAs/GaAs quantum-dot structures has been evidenced by means of a single-dot spectroscopy study. Also the effect on the charge state of the dot is demonstrated to be markedly dependent on the carrier transport and capture into the dot. By means of the essential tool in the regulation of the carrier transport, a tunable lateral electric field, it is demonstrated that the charge state accumulated in a single QD and its emission efficiency can be controlled. It is established that the potential fluctuations in the wetting layer play an essential role in localizing, in particular, the heavier holes. Accordingly, the charge state of the dot is essentially due to the field-induced tunneling of the holes out from these localization potentials, to subsequently become captured into the dot, and accordingly effectively neutralizing the charge configuration of the dot.

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[26] For our calculations we have employed an in-plane hole effective mass of $m^*=0.155m_0$ [J. Brübach, A. Y. Silov, J. E. M. Haverkort, W. v. d. Vleuten, and J. H. Wolter, Phys. Rev. B **59**, 10315 (1999)] for the case of ultrathin InAs quantum wells embedded between GaAs barriers. $F$ is calculated as $0.65V_{dc}/(15\ \mu m)$, where the factor of 0.65 accounts for the electric field reduction inside the structure.