Optical control of the exciton charge states of single quantum dots via impurity levels

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(Received 10 August 2005; revised manuscript received 20 September 2005; published 12 December 2005)

The impact of residual impurities on neutral and charged exciton complexes in single InGaAs quantum dots (QDs) grown by metalorganic chemical vapor deposition were investigated by microphotoluminescence combined with photon correlation measurements. We show that the formation of a charged exciton can be controlled by using resonant excitation to the residual impurity level. This optical excitation scheme is useful for the selective generation of only charged excitons in initially neutral QDs without sophisticated sample designs.

DOI: 10.1103/PhysRevB.72.233302 PACS number(s): 73.21.La, 78.67.Hc, 71.35.−y, 78.55.Cr

Optical spectroscopy of single semiconductor quantum dots (QDs) has recently been the subject of intensive investigation.[1–13] This technique not only provides direct access to the underlying atomlike properties of single QDs, but also opens up possibilities of utilizing QDs as building blocks for quantum information applications, such as single photon emitters[14–17] and quantum logic gates.[18–20] For many of the proposed applications, few-particle exciton states of single QDs are of great importance. The few-particle exciton states arise from different charge (electrons and holes) configurations in a QD, including neutral excitonic species as well as their charged counterparts. Of particular interest are singly, charged excitons (or trions), which are free from the electron-hole exchange splitting due to their singlet spin states[11,21–23] and hence are well suited for single-photon emitters without fine structures.[17] In addition, the single spins left after trion recombination are also useful for spin-based quantum information processing.[24,25] In this respect, controlling the QD charge state appears to be an important step toward these applications.

The QD charge states can be electrically controlled using charge tunable structures[9,10,22–25] to load extra charges sequentially from the back contact by tuning the gate voltage. Optical control[4–6,8,12,13] is also feasible, but is usually based on more sophisticated mechanisms and/or sample designs. Because the photogenerated carriers are neutral in nature, the formation of charged excitonic species relies on the capture of an unequal number of electrons and holes into a QD. Two different approaches have been proposed to achieve this goal. The first utilizes QDs that are initially filled with a number of electrons from its *n*-doped surroundings and exploits the photodepletion effect[4] to control the QD charge states. This approach has also been extended to further utilize coupled quantum wells for creating spatially separated photogenerated electrons and holes.[5] In both cases, the optical excitation intensity can be used to tune the QD charge state from negative to positive configurations.[4,5] The second approach is based on initially empty QDs.[12] The main idea is to optically inject electrons and holes pairwise into the QDs, together with excess electrons from acceptor impurities, preferentially forming negatively charged excitons. Dual laser excitation schemes[13] have also been proposed, making possible optically gated control of the exciton charge states.

While optical charging of QD has been reported in the above studies, the identification of different spectral lines is still a fundamental issue to be addressed. In fact, investigations based solely on microphotoluminescence (μPL) studies are inadequate to rule out the possibility that some emission lines may arise from different dots. Furthermore, the rich line structures in single QD spectra could arise not only from the multiparticle interactions, but also from the interactions between QDs and charged impurities and or defects nearby, which may further give rise to an exciton peak multiplet.[7]

In this work, we use the μPL technique combined with photon correlation measurements to investigate the neutral and charged exciton complexes in single InGaAs QDs grown by metalorganic chemical vapor deposition (MOCVD). The impact of residual impurities are expected to be more significant in MOCVD-grown samples and hence are addressed in the present study. Photon cross-correlation measurements were used to unambiguously identify different exciton lines.[26–29] We show that the formation of charged excitons can be controlled by using resonant excitation to the residual impurity level. This optical excitation scheme is useful for selective generation of only charged excitons in initially neutral QDs without sophisticated sample designs. The fine structures caused by QD-impurity interactions are also addressed.

Our sample was grown on a n^+ GaAs substrate by a low-pressure MOCVD reactor using trimethylgallium (TMGa), trimethylindium (TMIn), and arsine (AsH_3) as source materials. After a 500-nm undoped GaAs buffer layer, a layer of $In_{0.5}Ga_{0.5}As$ QDs was then grown at 500° C. The InGaAs coverage was carefully controlled to ~4.2 MLs, yielding a low QD density of about 10^8–10^9 cm^{-2}. The QDs were finally capped by a 80-nm undoped GaAs layer. Further isolation of individual QDs was achieved by fabricating an aluminum shadow mask with an array of 300-nm apertures using electron-beam lithography. μPL was performed at 5–8 K on these apertures using a Ti:sapphire laser focused via a microscope objective (numerical aperature=0.5). Luminescence was collected by the same objective lens and analyzed by a 0.64 m triple monochromator equipped with a charge-coupled device (CCD) camera. The typical integration time is 60 s. Photon correlation measurements were performed using a Hanbury-Brown and Twiss (HBT) setup,[14–17] consisting of a beam splitter and a pair of Si avalanche photodiodes (APDs). For cross-correlation measurements,[26–29] we inserted a monochromator (resolution ~0.5 nm) into each arm

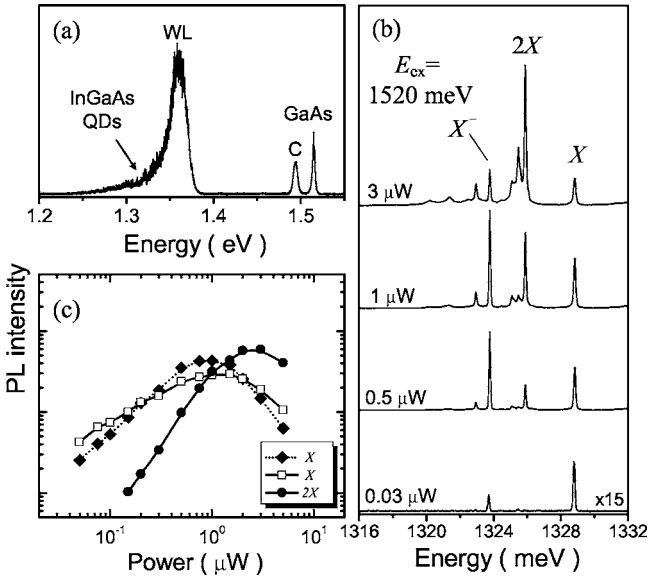

FIG. 1. (a) The μPL spectrum of the unmasked regions. (b) The power evolution of single QD spectra for $E_{ex}=1520$ meV. (c) The intensities of X, $2X$, and X^- lines as a function of the excitation power.

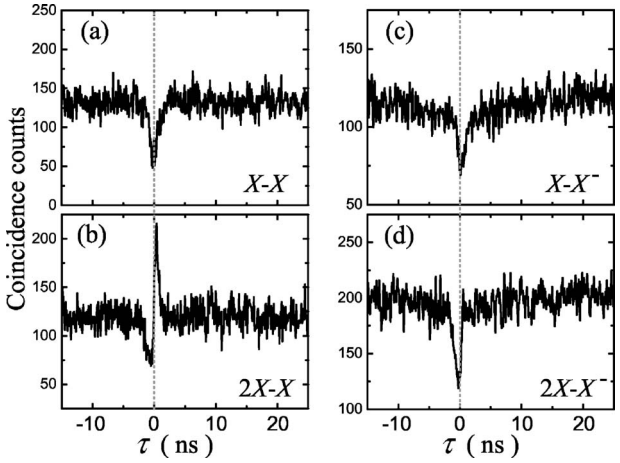

FIG. 2. Autocorrelation of X line (a) and cross-correlations obtained by using $2X$ and X lines (b), X and X^- lines, (c) as well as $2X$ and X^- lines, and (d) as start and stop triggers.

for spectrally selecting different lines. The outputs of the two APDs were sent into a time-correlated photon counting card for recording the histograms of collected coincident counts as a function of time delay τ, which is equivalent to the second-order correlation function, $g^{(2)}(\tau)$, after normalization.

Figure 1(a) shows the μPL spectrum of the unmasked regions. Apart from the GaAs band-edge emission at 1.519 eV, an impurity related peak at 1.494 eV is also observed. The most likely impurity is carbon, which is the kind of residual acceptor commonly seen in MOCVD-grown samples. The 1.363-eV peak is the wetting layer (WL) emission. Due to the low QD density, the QD signals emerge as a tail consisting of a series of spectral lines below the WL peak. Individual QD spectra can be accessed through the fabricated apertures. Several apertures containing only one QD were investigated, all of which showed similar behavior. Figure 1(b) shows the μPL spectra obtained from one of the apertures as a function of excitation power (P_{ex}) for an excitation energy of $E_{ex}=1520$ meV. These spectra display a number of competing lines, corresponding to various multiparticle configurations in the QD. The emission lines at 1328.8 meV and 1325.9 meV are identified as single exciton (X) and biexciton ($2X$) lines, respectively, according to their linear and quadratic power dependence of intensity, as shown in Fig. 1(c). Apart from the X and $2X$ lines, a prominent line was also observed at 1323.8 meV, separated from the X line by -5 meV. This line showed a power dependence very similar to that of the X line, hence it was ascribed to the single negatively charged exciton (X^-).[30]

The assignment of these competing lines can be further confirmed by photon cross-correlation measurements.[26–29] We first measured the autocorrelation of the X line under continuous-wave and above-band ($E_{ex}>1520$ meV) excitation, which is shown in Fig. 2(a). It displays a symmetric antibunching dip at $\tau=0$, with a normalized depth of $g^{(2)}(0)=0.38$, indicating that the emitted photons originate from a single quantum emitter. The cross-correlations between the $2X$ and X lines, the X and X^- lines, as well as between the $2X$ and X^- lines, are shown in Figs. 2(c) and 2(d), respectively. For the $2X$-X cross-correlation, both bunching ($\tau>0$) and antibunching ($\tau<0$) are displayed, which can be considered as the characteristic feature of a cascade process for the $2X$-X photon pairs.[26,27] As for the X-X^- and $2X$-X^- cross-correlations, both correlation curves feature an asymmetric antibunching dip, inferring that these lines indeed do arise from *the same* QD, but from different excitonic species. For example, the longer recovery time for $\tau>0$ in the X-X^- cross-correlation confirms that X^- is a trion state because of the relatively shorter (longer) time scale required for recapturing one hole (two electrons and one hole) to form an $X(X^-)$ state after the emission of an $X^-(X)$ photon.[27] The asymmetric antibunching dip in the $2X$-X^- cross-correlation can also be explained in the same way. Besides, we noted that the $2X$-X^- cross-correlation doesn't show bunching behavior for $\tau>0$, i.e., the emissions of $2X$ and X^- photons are not sequential. It has been reported that $2X$-X^- photon pairs can be sequential emissions if the time scale for recapturing one electron is faster than (or at least comparable with) the X recombination lifetime.[28] In our case, this recapturing time seems to be not fast enough to initiate such cascade $2X$-X^- emissions.

The formation of charged excitons was usually related to the presence of impurities in the QD surrounding. For the MOCVD-grown sample used here, the unintentionally doped carbon impurities may play a prominent role. To study the impurity effects, we investigated the QD spectra using different E_{ex}'s, which are shown in Fig. 3(a). For $E_{ex}=1520$ meV, both the X and X^- lines appear in the spectrum. As E_{ex} is reduced to (or even below) the WL energy ($E_{WL}=1363$ meV), only the X line can be observed. This implies that the QD is initially *neutral* in the absence of laser excitation. Since the QD layer is close to the surface, the QDs are empty and the residual carbon acceptors are ionized by the surface electric field. Under above-band excitations,

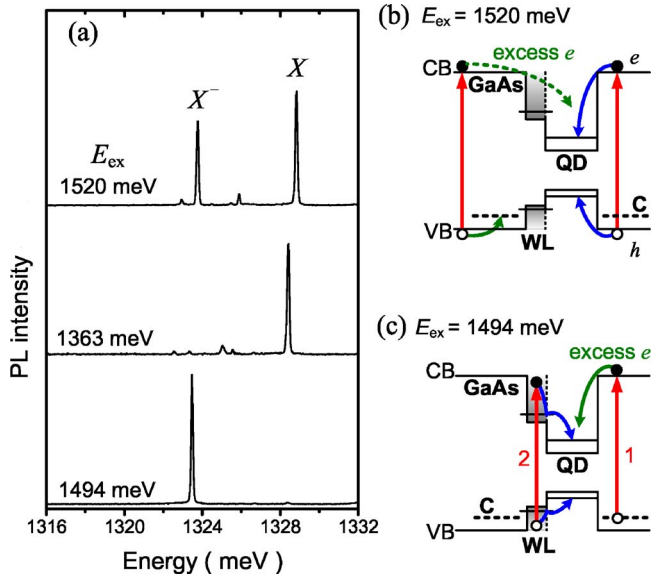

FIG. 3. (Color online) (a) Single QD spectra under different E_{ex}'s. (b) and (c) The optical processes for above-band excitation and resonant excitation to the carbon acceptor level.

the electrons and holes may be captured either pairwise or separately into the QD, forming either neutral or charged excitonic species. The carbon acceptors here act as hole traps around the QDs [see Fig. 3(b)], hence the formation of $X^-(X^+)$ is favored (unlikely). However, the steady state occupation of either X^- or X still depends on the transport properties of photogenerated carriers in the GaAs barrier or the WL,[6,8] selective generation of only one exciton type is difficult for above-band excitations. Of particular interest is that when $E_{ex}=1494$ meV, i.e., the resonant energy of carbon acceptor level, selective generation of only X^- becomes possible. This can be explained by the optical processes depicted in Fig. 3(c). The resonant laser light can be absorbed by the carbon acceptors, and they inject free electrons into the GaAs conduction band, while leaving bound holes in these acceptors [see process 1 in Fig. 3(c)]. In addition, part of the laser light can also be absorbed by the WL, thereby creating electrons and holes pairwise in the WL [and subsequently into the QD, see process 2 in Fig. 3(c)]. Since most of the photogenerated holes are trapped by the carbon impurities, the formation of neutral X is very unlikely, leading to the preferential generation of only X^- in the QD.

The optical excitation scheme, based on resonant excitation to the impurity level, allows us to selectively excite only X^- in neutral QD. This behavior is illustrated in Fig. 4(a), where the power evolution of the single QD spectra for $E_{ex}=1494$ meV is displayed. The spectrum is dominated by the X^- line in the low P_{ex} regime. As can be seen from Fig. 4(b), the X^- line exhibits a saturation power, P_{sat}, at 30 μW, above which the neutral exciton lines (X and $2X$) become increasingly visible. Since the formation of X^- relies on the injection of excess electrons from the residual carbon impurities, the intensity saturation means that the impurity absorption process has been saturated by the photogenerated holes. Therefore, a further increased P_{ex} can no longer inject excess electrons from the fully occupied carbon acceptors, making the QDs more and more neutral for $P_{ex}>P_{sat}$. This behavior can also be inferred from the spectrum with $P_{ex}=100$ μW, where the spectral feature becomes similar to those using above-band excitations shown in Fig. 1(a).

The emission energies of the X^- and X lines (E_{X^-} and E_X) also varied with P_{ex}, which are shown in Figs. 4(c) and 4(d),

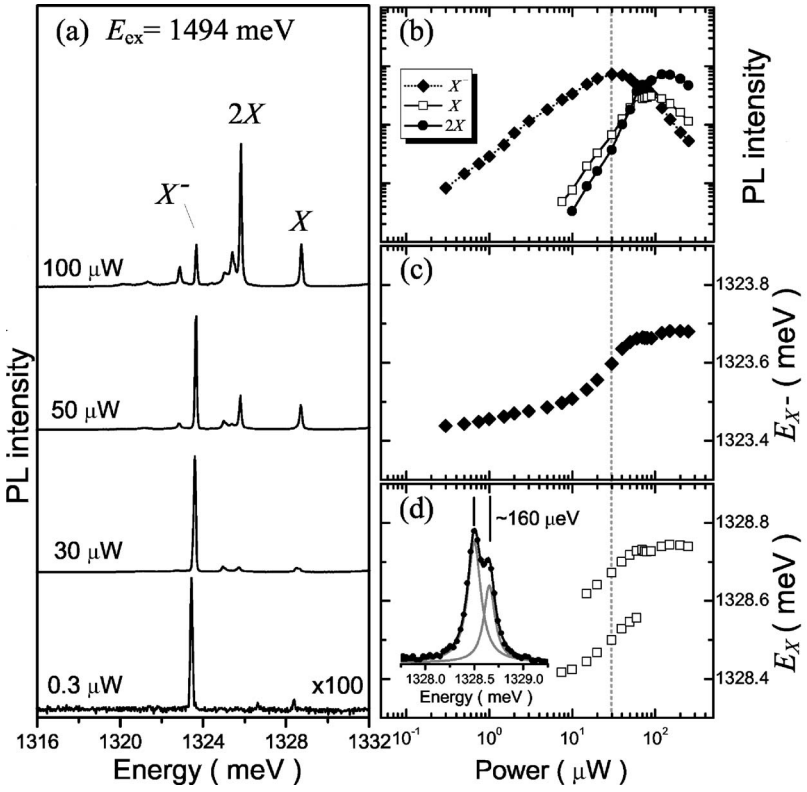

FIG. 4. (a) The power evolution of a single QD spectra for $E_{ex}=1494$ meV. (b) The intensities of X, $2X$ and X^- as a function of excitation power. (c),(d) The energies of the X^- and X lines as a function of excitation power. P_{sat} of the X^- line is indicated by a vertical dotted line. The inset in (d) shows the X peak doublet at P_{sat}.

respectively. We found that E_{X^-} displays a sigmoid energy shift and an abrupt change at P_{sat}. For the case of E_X, it further shows a peak doublet near P_{sat}, exhibiting a fine structure splitting of about ~ 160 μeV [see the inset of Fig. 4(d)]. Since the peak doublet did not show any polarization correlation, this energy splitting cannot be ascribed to the electron-hole exchange splitting. In addition, no exchange fine structure is expected for the trion state because the two electrons of X^- are paired in a singlet configuration. Therefore, we ascribe the X peak doublet, as well as the sigmoid shift in X^-, to the interactions between the QD and charged impurities nearby. A similar effects have been reported in Ref. 7, in which a multiplicity of X lines were observed where the impurities and/or defects around a QD are randomly neutralized and ionized by the photogenerated carriers under the below gap excitations. In our case, the observed energy shifts, in accordance with P_{sat}, give clear evidence for this effect. For $P_{\mathrm{ex}} < P_{\mathrm{sat}}$, most of the residual impurities are ionized. If there is a carbon acceptor located near the QD, the ionized acceptor atom will give rise to an electric field and, hence, to Stark shifts in emission energies for these exciton states. Under a higher P_{ex}, such a charged impurity can be neutralized by the photogenerated carriers, thereby eliminating the Stark shifts. Therefore when P_{ex} was varied across P_{sat}, an abrupt change in emission energy, or even a peak doublet, may be observed near P_{sat}. However, it is not possible to estimate this energy shift quantitatively since the location of the impurity relative to the QD is not known exactly. Nevertheless, the observed splittings are typically <200 μeV, which implies that the carbon impurities may be at least a few tens nanometers away from the QDs.

In summary, we have applied μPL and photon correlation spectroscopes to study the neutral and charged exciton complexes in single InGaAs QDs grown by MOCVD. We show that the residual carbon acceptors, commonly seen in the MOCVD-grown sample, can act as mediums for selective generation of only charged excitons in initially neutral QDs by using resonant excitation's to the impurity level. We also observe fine-structure splittings induced by the QD-impurity interactions, which should be considered for further applications.

This work was supported by the National Science Council of the Republic of China under Grant No. NSC-94-2120-M-008-001.

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