Photoluminescence of single GaN/AlN hexagonal quantum dots on Si(111): Spectral diffusion effects

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We report microphotoluminescence studies of single GaN/AlN quantum dots grown by molecular beam epitaxy on Si(111) substrates. Small groups of emission lines characterize each single dot, with linewidths mostly limited by our experimental setup to 1 or 2 meV. By using time-dependent microphotoluminescence, we observe both the continuous and discontinuous spectral diffusion of these lines, assigned to the trapping of charges at defects in the vicinity of the dots. We show that this trapping takes place on a large variety of time scales, which depend on the photogeneration. It induces energy shifts that also cover some range, yielding both some unresolved broadening and discrete positions of the emission line. We propose that this results from different local configurations, mainly in terms of the distance between the defects and the dots.

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I. INTRODUCTION

Among the variety of low-dimensional semiconductor nano-objects, novel quantum dots (QDs) based on group III nitride compounds have recently received some attention. Within the context of efficient light emitting devices in the visible and ultraviolet ranges of the electromagnetic spectrum, these QDs are promising candidates for the realization of solid state single photon sources operating at room temperature. In particular, GaN/AlN QDs are expected to provide satisfactory confinement of electron-hole (*e-h*) pairs, due to a band gap difference larger than 2 eV between GaN and AlN. One key issue, when one aims at using QDs as controlled single photon emitters, is the broadening of the luminescence lines of single QDs, as measured by microphotoluminescence (μPL) or cathodoluminescence (CL). Indeed, the spectral separation between the exciton and biexciton lines has to be larger than this broadening, so that the single exciton recombination can be isolated.[1,2] Moreover, such a decoherence mechanism has to be controlled if one intends to use these systems for quantum information processing.[3,4]

Generally speaking, the major source of broadening of single QD sharp luminescence lines is the coupling to phonons[5–7] yielding side wings on the lines that develop when increasing the sample temperature. Other environmental effects also play a significant role, such as the so-called "spectral diffusion" effect induced by the trapping of free carriers at defects in the vicinity of the QDs. This spectral diffusion manifests itself by small, time-dependent energy shifts of the line, as a result of some Stark effect.[8–11]

In the field of nitride QDs, spectral diffusion has been only reported and studied, so far, on (Ga,In)N QDs, by CL and μPL.[12,13] This effect has also been mentioned as the origin of the large (~10 meV) inhomogeneous broadening of single-dot emission lines measured by μPL on GaN/AlN QDs grown by metal organic chemical vapor deposition on the (0001) surface of a 6H-SiC substrate.[14] As a matter of fact, only a few μPL results have been published so far on GaN/AlN single QDs.[15] The only other example is that of QDs grown by molecular beam epitaxy on the nonpolar (11–20) face of 6H-SiC substrate.[16] This choice of growth plane aims at reducing the effects of the huge internal electric field that normally exists perpendicular to the growth plane, for wurtzite GaN QDs grown along the (0001) axis. Indeed, a field as large as 9 MV/cm can be present, inducing a giant quantum-confined Stark effect, red shifting the ground-state optical transition, and drastically reducing its oscillator strength. In practice, for QD heights larger than 1.8 nm or so, the emission takes place at lower energy than the GaN band gap, with radiative lifetimes that grow nearly exponentially with QD height,[17] reaching milliseconds for 3.5 nm of QD height. This is not the case when QDs based on cubic GaN are grown:[18] the suppression of polarization fields yields nearly constant lifetimes of the order of 0.3 ns. By CL measurements on single cubic QDs, inhomogeneous broadening of 2 to 8 meV has been obtained.[19]

In this paper, we present μPL studies of single, polar GaN/AlN QDs grown on Si(111) substrates by molecular beam epitaxy. We observe sharper transition lines by μPL (~1.5 meV linewidth, limited by the setup resolution) if compared to previous works. We emphasize in particular, by time-dependent μPL analysis, the effects of spectral diffusion that allow us to identify multiple lines pertaining to the same single QD. In relation to their temporal characteristics, possible mechanisms for these spectral diffusion effects are discussed.

II. EXPERIMENT

We present single dot spectroscopy from two samples both consisting of a single plane of hexagonal GaN/AlN self-assembled QDs, obtained by Stranski-Krastanov growth mode transition, as shown in Fig. 1. Molecular beam epitaxy was used, allowing for the growth of GaN QDs along the (0001) axis, directly on top of a 400 nm AlN epilayer itself deposited on the (111) face of a Si substrate. Sample 1 was grown on Si substrate with a misorientation angle of 5°, inducing a moderate QD density of 10^{10} cm^{-2}, and was capped by a 50 nm thick AlN layer. Macro-PL spectra ex-

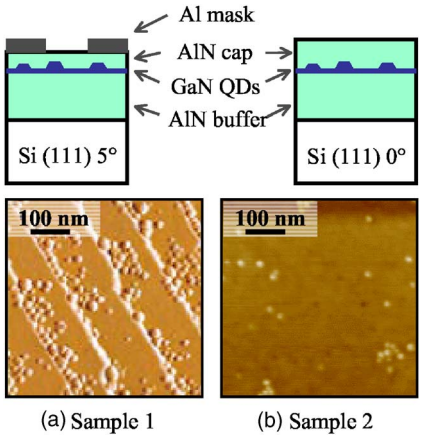

FIG. 1. (Color online) Descriptions and AFM images of the samples. (a) Sample 1 presents terraces since it is grown on 5° tilted Si(111). It has a moderate QD density, a 50 nm AlN cap, and an Al mask with 500 nm apertures. (b) Sample 2 presents a density gradient, a 175 nm AlN cap, and no mask.

hibit a bimodal distribution of the QD heights, h, with two populations: one around $h=2.8$ nm (peak emission at 3.2 eV) and a second one around $h=1.6$ nm (3.7 eV). In order to isolate a few QDs by μPL, this sample was covered with an aluminum mask in which apertures of 0.5 μm in diameter were opened by nanosphere photolithography.[20] Given the QD density, the number of QDs that we excite and from which we collect PL, through each aperture, is of the order of 100. Sample 2 was grown on a nominal (111) Si surface (no misorientation), but it presents a gradient of QD density over the sample, so that this density can be much smaller than for sample 1, around 10^9 cm^{-2} as shown in atomic force microscopy (AFM) images [Fig. 1(b)]. Therefore, this sample did not necessitate the use of any aluminum mask with apertures for performing the spectroscopy of isolated QD. The AlN cap for sample 2 has 175 nm of thickness. Macro-PL studies demonstrated the presence of three distinct populations in h, giving rise to rather narrow (250 meV) lines, spectrally separated, at 3.2 eV ($h=2.8$ nm), 3.6 eV (1.5 nm), and 4.2 eV (about 1 nm). In a previous study on similar samples,[17] we have measured radiative lifetime of 10 ns for QDs emitting at 3.5 eV (sample 1), and of 7 ns for QDs emitting at 3.7 eV (sample 2), respectively.

Our μPL setup allows for studies at low temperature (10 K), by using a cryostat with a continuous flow of liquid helium and an enhanced mechanical stability. We used a 36× microscope objective to focus the laser source to a spot diameter of ~1.5 μm on the sample surface. The μPL was excited by the frequency-doubled radiation of an ionized argon laser, at 244 nm (5.07 eV), in continuous-wave regime. This point is crucial for the following study, because it means that our laser creates e-h pairs only in the wetting layer or in the QDs, but not in the AlN barriers (6.2 eV of band gap). For the detection, we used a cooled charge-coupled device (CCD) camera, coupled to a 60 cm spectrometer with a 1200 grooves/mm grating. The entrance slit of the spectrometer had to be adjusted in order to preserve a satisfactory signal-to-noise ratio, yielding a spectral resolution comprised between 1 and 2 meV.

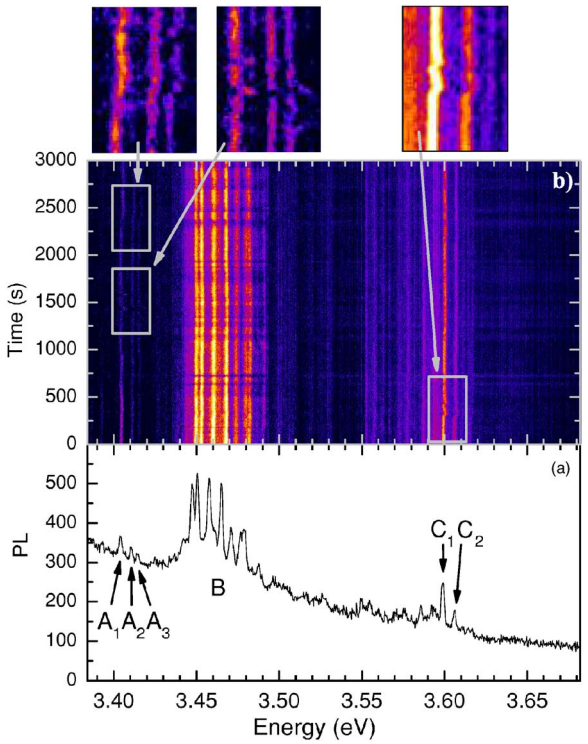

FIG. 2. (Color online) (a) Time-averaged μPL spectrum collected on sample 1, with an excitation density of 2 kW cm^{-2}; (b) time evolution of this spectrum over 3000 s with 10 s accumulation steps. The spectral resolution is 2 meV.

III. RESULTS AND DISCUSSION

Figure 2(a) shows one example of μPL spectrum of sample 1, resulting from signal accumulation over 3000 s. The excitation power density was about 2 kW/cm^2 in the focal plane of the microscope objective, and it could not be reduced because we needed to accumulate sufficient signal on reasonably small time intervals for the time-dependent analysis detailed below. Sharp emission lines originating from single QDs appear in the spectrum on top of a large and uniform background signal. We assign this background to the emission from other, larger QDs also present beneath the mask aperture, as well as neighboring QDs which are not discriminated by the mask. The emission lines in Fig. 2(a) can be *a priori* divided into subgroups of 2 to 7 lines, respectively, gathered around 3.40, 3.45, and 3.60 eV. Most of the emission lines of this spectrum have a full width at half maximum (FWHM) limited by the spectral resolution of 2 meV in this experiment, which is quite a general observation that we made for about 30 of such groups of lines, across the sample. Some lines are slightly broader: for C_1 and B_6 emission lines we have measured, respectively, a FWHM of 3 and 4 meV. Obviously, such linewidths are far beyond the values that one may expect for the homogeneous linewidth induced by interaction with the surrounding phonon bath.

At this stage, we are not able to ascertain that the above mentioned subgroups pertain to the same dot. In the time-averaged spectrum of Fig. 2(a), the distance between the lines in the same group is of the order of 6–7 meV (except

for the separation between B_1 and B_2 lines, 3 meV). Such separations are not easy to assign to a specific mechanism, among the variety of phenomena that may lead to multiple lines within a given dot.

Figure 2(b) displays a two-dimensional image showing the time evolution of the μPL spectrum of sample 1, the integration of which gave the spectrum in Fig. 2(a). This image is composed by the succession of 300 spectra taken on time windows of 10 s. The QD emission lines exhibit a time-dependent jitter (the so-called spectral diffusion) with an amplitude of about 2 meV. By following the emission line A_1 between $t=1200$ and 1800 s and between $t=2100$ and 2700 s, we can distinguish two types of spectral diffusion: the first one is discrete, with sudden jumps of the emission line between two clear-cut positions, whereas the second one is continuous, with the line slowly shifting over some spectral range. As previously reported for other systems,[12] we observe that some groups of 2 or 3 lines vary synchronously. For instance, the A_1, A_2, and A_3 lines exhibit the same spectral diffusion pattern. These synchronous phenomena have been already seen in II–VI semiconductor QDs and attributed to the local charge fluctuations in the vicinity of the QDs.[6] Clearly, the instants for which some shifts or jumps are observed are not the same for the lines pertaining to different groups. This means that the simultaneous behavior of some lines can be assigned to a common reaction to the *same* local electric field. Therefore we can now ascertain that the emission lines that have the same diffusion pattern arise from the same QD. This allows us to discriminate between at least three different QDs within the investigated spectral range. For instance, the A_1, A_2, and A_3 lines clearly correspond to a single QD; the same is true for the C_1 and C_2 lines. As for the *B* group, the spectral diffusion, if any, does not take place on time scales larger than seconds and therefore we cannot conclusively assign these lines to an unique QD.

Now the question has to be raised on the nature of the different lines that are observed for a given single QD. Previous works on II-VI QDs[11] reported detailed analysis based on the application of an external magnetic field and on the careful study of spectral diffusion phenomena. These works proved that the different emission lines resulted from the existence of a variety of excitonic complexes such as negative or positive trions that exhibit different energy shifts under the same perturbation induced by a given charge in the neighborhood of the dot.

The case of GaN/AlN wurtzite QDs is, *a priori*, much more complex, first of all because of the existence of a huge internal electric field that renders their situation quite different from all the systems previously studied. For example, the Coulomb correlation energy between the electron and the hole inside such a QD is extremely size dependent, because of the separation of the two types of carriers by the field. This energy is on the order of a few tens of meV but it can be reduced below the bulk exciton binding energy (27 meV) if the QD height is large enough. Unfortunately for the sake of modeling, this Coulombic energy is of the same order of magnitude as the expected splitting between states originating from the three-fold valence band in wurtzite GaN (about 10 meV in the bulk material). Moreover, similar orders of magnitudes (slightly larger, in fact) have recently been reported[21] for the energy separation between the single exciton and trion or biexciton complexes that appear systematically at higher energy than the exciton, due to the domination of repulsive Coulombic terms in the Hamiltonian. This domination results from the presence of the strong electric field that pushes charges of the same sign towards the same side of the QD, along the growth axis. Quantitatively, there might be a discrepancy between theoretical calculations and our experiments because we have recently proved[17] that the electric field in our samples (single QD planes) is close to that proposed in early theoretical predictions[22] and therefore much larger than the commonly accepted value,[23] which in fact corresponds to a measurement made on multiple QD planes, where the field is reduced.[22,24]

For this reason, we cannot discard, *a priori*, the possibility that the observed groups of lines correspond to differently charged excitonic complexes. In fact, we do not discard, either, the possibility that charged complexes could be formed between an exciton in a given QD and a single charge located in a neighboring QD, or even trapped on a charged defect, nearby (a donor, for instance). This may explain the relatively small energy splitting (6–7 meV) that we observed between the different lines, if compared to theoretical calculations of "intradot" excitonic complexes.[23,25]

In fact, a quite attractive explanation for the observed multiple lines is the one of trapped charges in the very close vicinity of the QD. As a matter of fact, recent studies have proved that the application of a longitudinal[26] or in-plane[14] electric field is able to shift the transition energy of a single QD by several meV. Whence the idea that a charged defect, distant by a few nanometers from the QD, may induce such discrete shifts of the fundamental, neutral exciton, with an amplitude controlled by the distance that separates the defect from the dot. If the time necessary for the charging and/or emptying mechanism of the defect is much smaller than the elementary time window that we use (1 s or 10 s), then we can observe several configurations within the same time window. Here, it is important to remember that we create *e-h* pairs only in the luminescent system itself and not in the AlN barriers. This means that it is really those charged defects which are very close to (or even within) the system (wetting layer +QDs) that may induce discrete jumps of the excitonic line on time scales much smaller than seconds, by trapping and releasing carriers from and back to the system. Following this scenario, the continuous or discrete spectral diffusion that we show in Fig. 2 would be provoked by other charged traps, lying at a larger distance from the QD, or even by the presence and/or absence of *e-h* dipoles in the surrounding QDs.

To obtain an idea of the most likely possibilities, we can calculate the electric field induced, at the center of a given QD, by a fixed elementary charge, as a function of the distance between them. We find, for instance, that a point charge placed at 5 nm from the QD can induce an average field of $\sim$60 kV/cm in the dot. Referring to the recent works mentioned above,[14,26] we estimate that such a field can shift the excitonic transition by $\sim$8 meV, if the charge is placed vertically above or beneath the QD, or by $\sim$3 meV, if the charge is placed along the growth plane. In comparison, an *e-h* dipole placed in an identical QD situated at a distance of

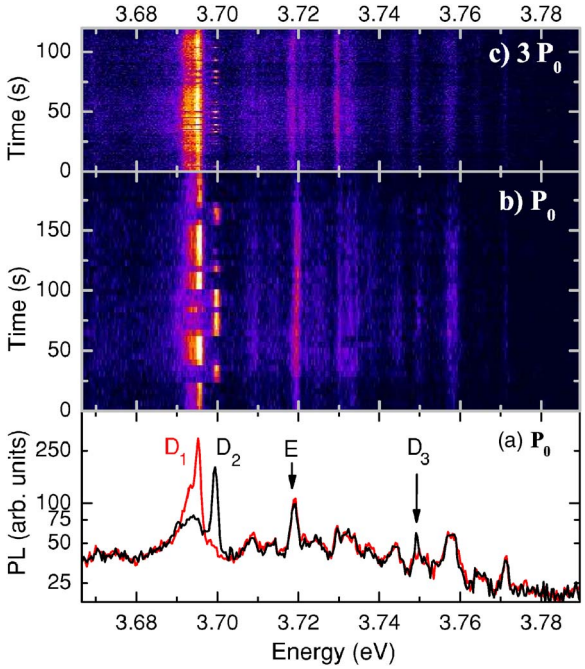

FIG. 3. (Color online) (a) Time-averaged μPL spectrum of sample 2 in the two observed configurations (see text) at low excitation density (P_0=230 W cm^{-2}) and on a logarithmic vertical scale. (b) Time evolution of the spectrum at the same excitation density P_0 and (c) at $3P_0$, with an accumulation step of 5 s, and 1 s, respectively. The spectral resolution is 1.5 meV.

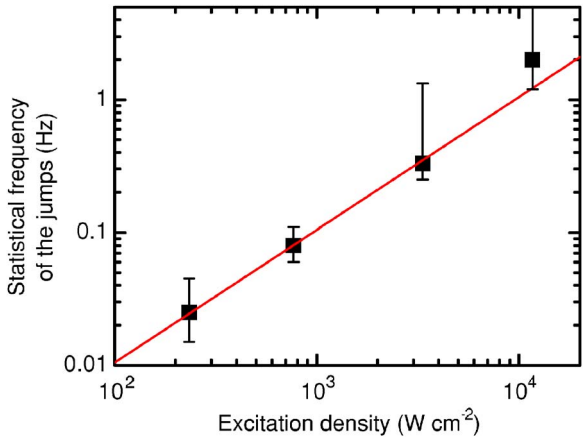

FIG. 4. (Color online) Statistical frequency of the jumps observed in Fig. 3(b). The linear regression curve is shown by the (red) line.

10 nm from the observed one (nearly the close-packed situation, considering the in-plane size of our dots), would only induce a longitudinal electric field of a few kV/cm, and therefore an energy shift of a fraction of a meV.

From these estimations, we conclude that the most probable scenario is the existence of some traps (e.g., donors) in the close vicinity of *some* QDs. There can be one or several traps, yielding several PL lines, such as for the case of the A_1, A_2, and A_3 lines in Fig. 1. The effect of other traps, at larger distances, would then be to slightly shift these discrete lines.

In fact, one important issue concerning the physics of those charged traps is the carrier lifetime on them. This lifetime can depend strongly on the position of the trap from the luminescent system and on the overall excitation conditions of the sample.

For example, Fig. 3 shows the μPL results obtained on a region of sample 2. For this sample, the QD density is much smaller than for sample 1 and, therefore, no metallic mask was used. We could also vary more easily the excitation power density and use shorter time windows and a narrower entrance slit, because the collection of the PL signal was more efficient without a mask. The most remarkable result in Fig. 3(b) is the observation of discontinuous jumps for a given line that "beats" alternatively between two positions separated by 4 meV, with duration of several tens of seconds at each position. We identified the time windows in which the main line is either at the position D_1 or at D_2, respectively. By averaging separately the PL signal measured over either type of time windows we obtain the spectra presented in Fig. 3(a). Clearly, all the minor, unlabeled PL lines and even some more important ones, such as the E line, do not vary at all and therefore do not correspond to the same QD as D_1 and D_2. As previously remarked, D_1 and D_2 are related to the same excitonic state in the presence and absence of a charge in neighboring traps, respectively. Quite interestingly, the above signal treatment allows us to identify another (weak) transition that *does* pertain to the same QD, labeled D_3 in Fig. 3(a). Lines D_2 and D_3 are time correlated: whatever the excitation power density, they appear and disappear simultaneously. They are separated by 50 meV. This value is of the order of magnitude of those theoretically predicted[23,25] for the energy separation between different excitonic complexes in GaN/AlN QDs. However, further investigations are needed in order to assess that the D_3 transition is related to such an excitonic complex.

Similar to the subgroups of lines in Fig. 2, we attribute lines D_1 and D_2 to the presence and/or absence of a charge on a very close trap. However, in the case of Fig. 3(b), the trapping occurs on a time scale which is longer than our acquisition time and, therefore, we can separate in time the electrostatic states that we could not separate for sample 1 (A_1, A_2, and A_3 lines, for example). In fact, the explanation for this difference is given by the study that we performed of these discontinuous jumps, for sample 2, as a function of the excitation power density, between 0.23 and 12 kW/cm^2. Figure 3(c) shows that the discrete, random shifting between the two emission lines occurs at a faster rate as the excitation density is increased. In Fig. 4, we show that the statistical frequency of the jumps depends linearly on excitation power density, in the investigated range. Therefore the trapping mechanisms as well as the lifetime of the charge on the trap are directly related to the photogeneration of e-h pairs in the wetting layer and in the QDs. These results suggest that the exchange rate of carriers between the traps and the QDs is a growing function of both the excitation power density and the proximity of the trap, which is scaled by the splitting induced on the exciton line.

If, as strongly indicated by the induced Stark shifts, the traps present a central electrostatic potential, we can readily

infer that the trapping rate decreases exponentially with the distance between the traps and the system. Given the splittings measured in sample 1 (6–7 meV) and in sample 2 (4 meV), and the power density used for the spectrum in Fig. 2 (2 kW/cm^2), we now understand why the different configurations could be observed simultaneously within the same time window of 10 s for sample 1, whereas we could separate them for sample 2: in the second case, we could use much lower power densities and the involved trap is probably a little further away from the QD.

Our results evidence two energy scales for the spectral diffusion and show that the charge fluctuations in the environment of each QD do not only lead to an inhomogeneous broadening of the transition lines. Such a broadening is observed when the typical fluctuation time is shorter than the acquisition time window but larger than the other decoherence times of the system, and when a large number of electrostatic configurations are accessible around the QD. In our samples, this inhomogeneous broadening is smaller than the one previously reported[14] and, in practice, it probably falls beneath the spectral resolution of our setup. Moreover, on the same energy range (10 meV), where a single, inhomogeneously broadened line was reported by another group,[14] we observe distinct transitions instead. These observations suggest that the local electric field can only take a few distinct values, i.e., that the density of traps in the close vicinity of the QD is much smaller in our samples. We may emphasize that the same effect is observed with and without an aluminum mask, and for a thin (50 nm) and a thick (175 nm) AlN cap layer. Thus, we do not believe that the observed spectral diffusion is related to the vicinity of the surface, but rather to the intrinsic properties of the QDs and of their electrostatic environment, in particular to the quality of AlN barriers.

Before concluding, we wish to remark that the production of multiple lines by the electrostatic environment, with energy splittings beyond the spectral broadening, has been reported for CdTe QDs,[6] but with much smaller amplitudes than those obtained here on GaN/AlN QDs. This effect has not been reported either, to our knowledge, for single InAs QDs. We believe that our observations are related to the polar character of our QDs, making their optical transitions more sensitive to the influence of neighboring charges, and also to the insulating properties of AlN, in which free carriers may be deeply trapped on long time scales.

IV. CONCLUSION

We have reported spectroscopic studies of single GaN/AlN QDs grown by molecular beam epitaxy on Si(111) substrates. We have observed linewidths mostly limited by our experimental setup to 1 or 2 meV, i.e., quite smaller than those previously reported in μPL studies. In particular, we have observed both the continuous and discontinuous spectral diffusion of the emission lines, assigned to the trapping of charges at defects in the vicinity of the QDs. We have shown that this trapping takes place on a large variety of time scales and produces effects that also cover a variety of energy scales. Its dynamics depends strongly on the photogeneration rate. We propose that this is the result of different local configurations, mainly in terms of the distance between the defects and the dots. Close charges give rise to different, discrete positions of the emission line within typically a range of 10 meV, whereas more distant charges induce a continuous shift and an inhomogeneous broadening, with amplitudes of few meV. The assignment of multiple transition lines demands a lot of care and will certainly require further investigations in the future, in relation with the appropriate modeling of excitonic complexes in such QDs.

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