Subsecond Luminescence Intensity Fluctuations of Single CdSe Quantum Dots

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Photoluminescence (PL) intermittency characteristics are examined for single quantum dots (QDs) in a CdSe QD sample synthesized at a slow rate at 75 °C. Although the PL quantum efficiency was relatively low (∼0.25), we noticed that the PL intensity of single CdSe QDs fluctuated on a subsecond time scale with short-lived "on" and "off" states. The subsecond PL intensity fluctuations of CdSe QDs are different from "on" and "off" PL blinking generally observed for QDs fluctuating on a millisecond to minute time scale. We characterized single QDs by identifying polarized excitations, topographic imaging using atomic force microscopy (AFM), and transmission electron microscopy (TEM). From analysis of the PL intensity trajectories from >100 single CdSe QDs, the average intermittency time was 213 ms. From the PL quantum efficiency, slow growth of QDs, intensity trajectory analyses, and previous reports relating surface trap states and PL properties of QDs, we attribute the subsecond PL intensity fluctuations of single CdSe QDs and short-lived "on" and "off" states to a high-density distribution of homogeneous surface trap states.

Introduction

Colloidal semiconducting quantum dots (QDs) have received considerable attention for their unique optical properties including extended UV−vis absorption, narrow emission bandwidth, brightness, and photostability.[1−6] Recent advances in surface modification coupled with unique properties made QDs potential tags for fluorescence visualization of structures and functions of biological systems.[7−10] However, intrinsic luminescence intermittencies ("on" and "off" blinking)[11−19] of individual QDs on a wide time scale from milliseconds to minutes make it difficult to use them in single photon logic devices and continuous monitoring of labeled biomolecules. Although single QDs and single molecules show fluorescence intermittencies, fluorescence spectral changes and intermittency of single molecules are useful for investigating sensitive environmental changes.[20−24] The fluorescence intermittencies of single molecules are widely used for disclosing dynamics associated with several structural and functional transformations. Generally, prolonged spectroscopic and microscopic investigations involving single molecules are difficult due to fast photodarkening. On the other hand, photostability of QDs is not yet widely useful in single dot applications, limited by their intrinsic blinking nature and less sensitivity to environmental changes. Developing nonblinking QDs, suppression of blinking, and reduction of blinking time scale are necessary for efficiently utilizing the superior luminescence properties of QDs.

Since the discovery of luminescence intermittency of single QDs by Bawendi and co-workers[17] in 1996, several steady-state and time-resolved spectroscopic investigations have appeared foreseeing the origin of the intriguing photoluminescence (PL) intermittency.[11−16,18,19,25−36] Time-resolved spectroscopic investigations and spectral-shift measurements of single QDs proposed inhomogeneous charge localization and recapture to account for the PL intermittencies.[12,16,25,28] From previous ensemble-averaged[4,26,29,31,33,35,37] and single dot[12,25,28,30] time-resolved spectroscopic investigations, an excited electron localized to a surface trap state (charged QD) likely produces a nonemissive QD ("off" state) or a QD with deep trap emission. In many cases the trap state nonradiatively deactivates to the ground state at different time scales, producing different "on"/ "off" periods depending on the distribution of surface trap states, relocation of charge carrier near the surface, or reorganization of charges in the environment. The mechanism relating surface trap states, PL decay, and intermittencies of single QDs is inevitable and was supported by several investigations including PL enhancement of QDs by surface passivation with different capping agents,[1,32,34,38,39] suppression of blinking of single QDs by surface thiol passivation[13] and incorporating on rough gold surfaces,[40] and identifying "on"−"off" events at different temperatures.[11] Furthermore, reduction of surface trap states, contributed by dangling bonds, in band gaps by controlled growth of QDs has been known to provide better PL efficiency to QDs.[41] Summarizing previous investigations relating the PL intermittency and surface defects, QDs with reduced surface defects are expected to show reduced blinking and high PL efficiency.

In the current work we identified subsecond PL intensity fluctuations of CdSe QDs without long-lived "on" and "off" states. The present observation of subsecond PL intermittency

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of CdSe QDs is different from previous "on"−"off" blinking of QDs fluctuating on a wide time scale. From time-correlated single-molecule video microscopy analyses of the intensity fluctuations of >100 single QDs, the average intermittency time was 213 ms. The PL intermittency and short-lived "on"−"off" states are attributed to a high density distribution of homogeneous surface trap states. Investigation of subsecond intermittency statistics is not only important as a case study; specifically, our observations show possibilities for obtaining QDs with different PL intermittency characteristics or nonblinking QDs for single dot applications.

Experimental Section

Materials and Methods. CdSe QDs used in the current work were prepared following a literature method[42] with minor modifications. Streptavidin-coated core−shell CdSe−ZnS QDs (QD-585) were purchased from Quantum Dot Corporation, Hayward, CA, and were used as a standard for comparing the luminescence properties of CdSe QDs. Although we compared the luminescence properties of CdSe QDs with QD-585, commercial QDs are relatively less toxic due to the presence of a ZnS shell and a polymer overlayer. Furthermore, the presence of linkers such as streptavidin, biotin, and antibodies on the surface of commercial QDs are useful for a variety of biological labeling applications. Indeed, the CdSe QDs reported in the current work lack a ZnS shell and a protective polymer coating, and may be hazardous for direct biological applications. Cadmium acetate dihydrate [$Cd(Ac)_2 \cdot 2H_2O$], tri-*n*-octylphosphine (TOP), tri-*n*-octylphosphine oxide (TOPO), and selenium shots were purchased from Aldrich. Analytical grade methanol, 1-butanol, and acetone (Wako, Tokyo) were used as supplied. Slide glasses (Matsunami, Tokyo) were thoroughly cleaned prior to sample preparation by sonication in aqueous NaOH solution (0.1 M), Milli-Q water (Millipore Corporation, Billerica, MA), acetone, and Milli-Q water, each for 15 min, and dried in a jet of nitrogen gas.

Samples for fluorescence microscopy were prepared by spin coating (3000 rpm) TOPO-capped CdSe solutions (0.05 nM) on clean slide glasses. Reference samples were prepared by spin coating a 0.05 nM QD-585 solution on slide glasses (QD-585 was diluted using a specified buffer provided with the sample). Samples for atomic force microscopy (AFM) and transmission electron microscopy (TEM) imaging were prepared by spin coating CdSe QD solutions (1 nM in 1-butanol) on freshly cleaved mica plates and incubating on a carbon coated copper grid, respectively.

Absorption and emission spectra were recorded using a spectrophotometer (Hitachi U-4100, Hitachi, Tokyo) and spectrofluorometer (Hitachi F-4500, Hitachi, Tokyo), respectively. AFM images were recorded using a Nanoscope III microscope (Digital Instruments, Santa Barbara, CA). Tapping-mode AFM images were collected in air, using ultrasharp (radius of curvature 10−15 nm) silicon nanoprobes (Digital Instruments, Santa Barbara, CA). The cantilevers used were ∼120 μm long with a spring constant of ∼20−100 N/m and a resonance frequency of 356 kHz. TEM images were recorded using a 300 kV JEOL JEM 3010 microscope.

Single Quantum Dot Video Microscopy. We recorded the fluorescence images and PL intensity trajectories of single CdSe QDs and QD-585 using a wide-field video microscopy system; a combination of an inverted optical microscope (Olympus IX70) equipped with a long-pass filter (550 LP), a 100× objective (Olympus UMPlanFl, NA 0.95), an image intensifier (Video Scope VS4-1845), and a CCD camera (Hamamatsu
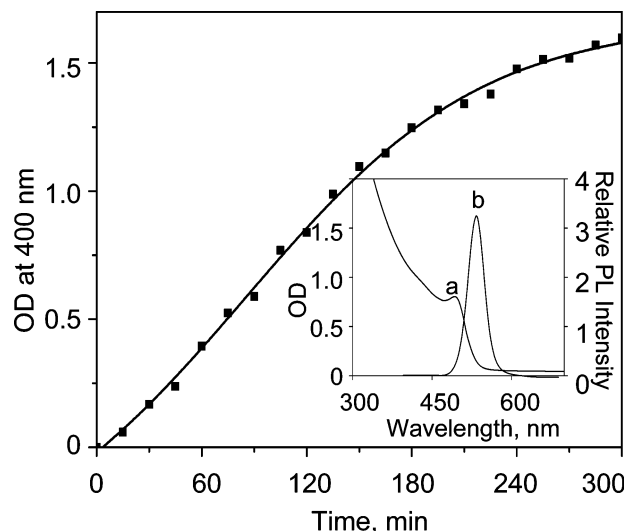

Figure 1. Temporal evolution of optical density at 400 nm during the reaction between $Cd(Ac)_2 \cdot 2H_2O$ and TOPSe at 75 °C. Inset: absorption (a) and PL (b) spectra of size-selected QDs.

C5985, for trajectory recording at 100 ms binning) was used. A second single photon counting (SPC) digital CCD camera (Hamamatsu C-4880-80) was used for recording trajectories at subvideo rates (20 ms binning). Excitation sources used for fluorescence imaging and trajectory recording were second harmonics (532 nm) of either a continuous wave Nd:YAG laser (Coherent DPSS 532) or a diode-pumped continuous wave Nd:YVO₄ laser (Spectra Physics Millennia IIs). Laser intensities used for imaging and trajectory recording were 17 and 2 W/cm², respectively. Single QDs were characterized by identifying the dipole orientation. For this, excitation polarization of a linearly polarized laser beam was changed following a literature method,[43] by rotating a polarizer in front of a quarter wave plate. We noticed no change of fluorescence intensity for fluorescent beads under similar experimental conditions.

Results and Discussion

Cadmium selenide QDs used in the current work were synthesized from $Cd(Ac)_2 \cdot 2H_2O$ and TOPSe, following a method developed by Peng and co-workers.[42] However, we adapted slow growth of QDs by lowering the reaction temperature. For a typical QD synthesis a round-bottom flask was charged with a mixture of $Cd(Ac)_2 \cdot 2H_2O$ (0.262 g, 1 mmol) and TOPO (3.86 g, 10 mmol), and heated at 75 °C for 30 min under continuous Ar purging. To this suspension 0.72 g of TOPSe (1 mmol) was injected in four scraps over a period of 20 min. TOPSe was prepared following a method described in the literature.[44] The reaction mixture was stirred at 75 °C under continuous Ar flow. Formation of CdSe QDs was identified from a gradual yellow coloration to the reaction mixture. Optical density at 400 nm recorded for aliquots of samples withdrawn from the reaction mixture at different time intervals is shown in Figure 1. We noticed a gradual increase of absorbance and PL intensity with time under reaction due to increase of CdSe concentration. However, essentially no spectral shift was observed with time under reaction. The reaction was allowed for 5 h and quenched by adding 10 mL of 1-butanol. The reaction mixture was cooled to 25 °C and centrifuged, and Cd-$(Ac)_2 \cdot 2H_2O$ left unreacted was removed. CdSe QDs were precipitated by adding 50 mL of methanol and isolated by centrifugation. A yellow residue of CdSe QDs obtained was purified by repeated precipitation from a 1-butanol:methanol

mixture (1:5 v/v), and provided 0.14 g (73%) of CdSe QDs. The CdSe sample contained QDs of different sizes with a major component emitting at ∼540 nm, identified from the PL spectrum. Narrow size dispersed QDs with PL maximum at 543 nm was isolated by two-stage size-selective precipitation from a mixture of 1-butanol and methanol (1:3 v/v). Centrifugation at 6000 rpm precipitated QDs of relatively larger size, identified from the disappearance of a long-wavelength component in the PL spectrum. Centrifugation was repeated at 10 000 rpm, during which the majority of QDs emitting ∼520−590 nm were precipitated, leaving smaller QDs emitting below 520 nm as major component in the solution phase. QDs precipitated in the second stage were collected and dissolved in a mixture of TOPO and 1-butanol (1:10 v/v). Typical absorption and PL spectra of the size-selected QDs are shown in Figure 1. We noticed essentially no contribution of deep trap emission of relatively low energy in the current work; likely the trapped electrons deactivated nonradiatively. The PL quantum efficiency of size-selected QDs was relatively low (ϕ_f ∼0.25) compared to QDs prepared at higher temperatures (ϕ_f = 0.4−0.8).[1,41,45] We determined the ϕ_f of CdSe QDs using a relative method employing Cy3 dye as a standard. The size-selected QDs were used for preparing single QD samples in the current work.

The slow rate of formation of CdSe QDs, noted from a gradual increase of absorbance in the UV−visible region, is likely responsible for subsecond PL intermittencies of single QDs. The gradual increase of optical density at 400 nm for equal amounts of samples withdrawn from the reaction mixture at different time intervals is shown in Figure 1. PL properties of QDs strongly depend on different factors such as growth temperature, amounts of precursor reagents, surface capping agents, reaction time, and sequence of addition of reagents.[1,8,41,42,44−46] Although surface capping of CdSe QDs using a higher band gap ZnS shell is a standard method for producing high-quality QDs,[1,46] several alternative synthetic strategies were also developed for producing high-quality QDs utilizing different precursor reagents and experimental conditions.[41,42,45] From different synthetic methods and investigations of PL properties of QDs, a set of parameters was identified for achieving high PL quantum efficiency. These include (1) surface passivation of QDs with different ligands, (2) preparation of shells on core QDs from materials of different band gaps, (3) photoinduced oxygen/ligand passivation of surface, and (4) controlled growth under optimum conditions. Reduced PL efficiency observed from time to time was attributed to surface electronic defects. Although PL efficiency was considerably increased by systematic investigations of different parameters involved in the synthesis of QDs, PL intermittency extending from milliseconds to minutes was retained for single QDs. Relatively low PL quantum efficiency (∼0.25) obtained in the current work is also likely due to surface defects. The quantum efficiency observed for CdSe QDs is not high compared to several recent literature reports on improving quantum efficiencies up to 60−80% by tailoring surface defect related properties. The proposed high density trap state model is supported by (1) recording and analysis of photoluminescence trajectories of >100 single quantum dots, (2) fluorescence quantum efficiency measurements at ensemble level, and (3) literature reports correlating trap states, intermittency, and quantum efficiency. Current work is different from previous reports on the synthesis and properties of CdSe QDs in that (1) the temperature (75 °C) of QD synthesis was relatively low compared to standard QD synthesis (200−300 °C), (2) the temporal evolution of size was not considerable at 75 °C, and (3) unusually fast PL intermit-

tencies and short-lived "on"−"off" states were observed for single CdSe QDs. We obtained insight into the present observation of subsecond PL intensity fluctuations of single CdSe QDs based on two important findings in the field of QD research: (1) PL intermittencies of QDs are related to surface defect states[4,11−13,15,16,28] and (2) surface defects are dependent on QD synthetic conditions and capping agents.[13,32−34,37,41,45] A systematic investigation including different parameters such as the temperature, amount of precursor reagents, reaction time, and type and amount of coordinating ligand is necessary for specifically characterizing a relation between intermittency and synthesis. We have synthesized CdSe QDs at three temperatures, 25, 50, and 75 °C. Considering that the melting temperature of TOPO is >60 °C, it was not used during the syntheses at 25 and 50 °C. We noticed relatively low PL quantum efficiencies of QDs synthesized without TOPO, likely due to aggregation. A direct comparison of PL properties among QDs synthesized at different temperatures was difficult considering the difference in the surface coordination (aggregates were always present in the absence of TOPO) and different PL quantum efficiencies. The present observation of subsecond PL intermittency shows future possibilities for developing nonblinking quantum dots by a systematic investigation of different synthetic parameters.

We used single-molecule video microscopy for recording and analyzing fluorescence intensity trajectories of single CdSe QDs. In air, individual CdSe QDs excited with a 532 nm laser showed fast intensity fluctuations on a subsecond time scale. Typical PL intensity trajectory of a single CdSe QD obtained with a temporal resolution of 100 ms is shown in Figure 2A. Details of PL intensity fluctuations are shown in a zoom-in trajectory in Figure 2B. PL intensity fluctuations observed for CdSe QDs are distinctly different from blinking of QDs reported previously in that long-lived "on" and "off" states were absent in the present case. Furthermore, PL intensity fluctuations of QDs are known to vary from dot to dot (static inhomogeneity) and also from time to time (dynamic inhomogeneity). In the current work we noticed essentially no static and dynamic inhomogeneities of intensity fluctuation. The validity of nearly homogeneous two-state intensity fluctuation of CdSe QDs even on a short time scale is presented with a 20 ms trajectory (Figure 2C). For comparison, the blinking characteristics of a standard QD-585 are shown in Figure 2D. Luminescence of QD-585 fluctuated on a wide time scale extending from milliseconds to minutes. All the trajectories were recorded under identical experimental conditions to avoid the possible influence of temperature and laser intensity on the PL properties of QDs. The contribution of background noise to the PL intensity fluctuations was not significant, based on PL intensity measurements of several CdSe QDs and QD-585 under different excitation powers. We have considered and investigated different possibilities for the subsecond PL intermittency characteristics and short-lived "on"−"off" states of single CdSe QDs, which are different from the PL properties of standard QDs. Transient changes of transition dipole orientation of a QD are likely to produce unusual PL intensity fluctuations in trajectories. However, in the present case the use of unbiased (circularly polarized) laser excitation ruled out a transition dipole effect in the subsecond PL intensity fluctuations of single CdSe QDs. Yet other possibilities— inclusion of more than one QD in the measuring area or the presence of QD aggregates—are also likely to contribute to unusual PL intensity fluctuations. We examined these two possibilities by identifying polarized excitations of single dots, and by AFM and TEM imaging of CdSe QD samples from solutions of different concentrations.
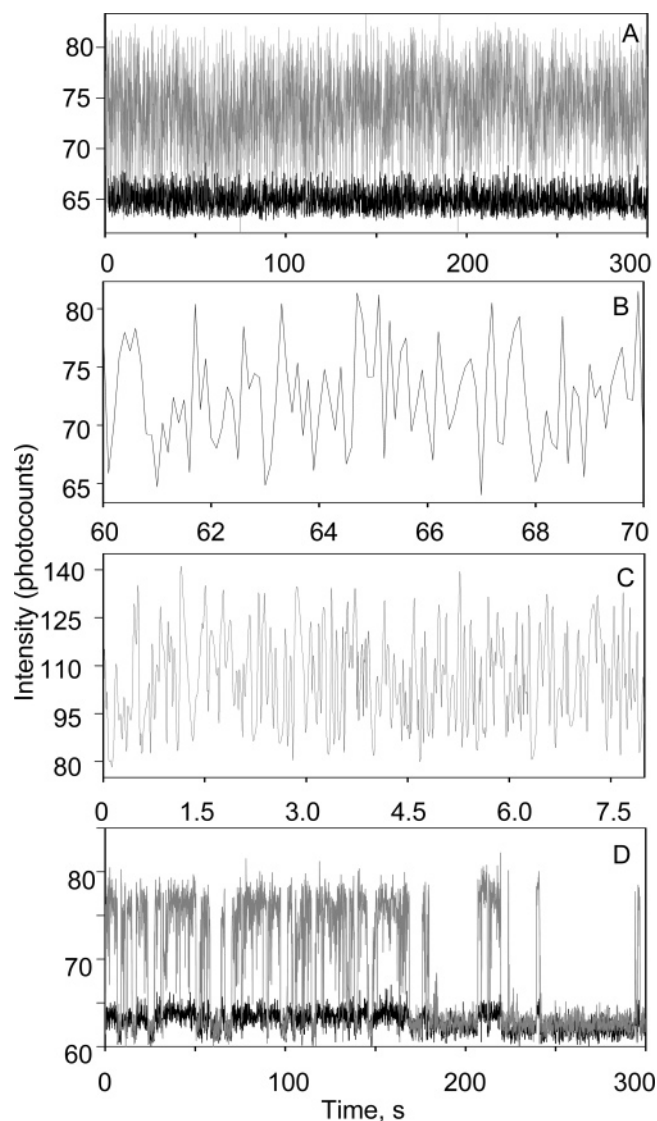
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Figure 2. PL intensity trajectories collected in air for single QD samples prepared by spin coating 0.05 nM solutions on slide glasses: (A) trajectory of a single CdSe QD with 100 ms binning, (B) a zoom-in trajectory of a single CdSe QD showing trajectory details of PL intensity fluctuations, (C) trajectory of a single CdSe QD with 20 ms binning showing two-state intermittency events even at short time scale, and (D) trajectory of a standard QD-585 with 100 ms binning. Trajectories were recorded under circularly polarized laser (532 nm) excitations. Excitation powers used were 2 kW/cm^2 for trajectories (A) and (D) and 10 kW/cm^2 for trajectory (C).

We identified polarized orientations of individual CdSe QDs from fluorescence images recorded under linearly polarized laser excitations of different polarization angles. Different methods, including photon antibunching,[47] identifying dark axis from emission polarization,[43] and identifying dipole orientation from excitation polarization,[48,49] are standard for characterizing single emitting moieties. In the current work we examined polarized excitations of CdSe QDs for characterizing single dots for technical reasons. We noticed that the PL intensity of CdSe QDs excited with a circularly polarized laser beam (532 nm) was not varied considerably from time to time and from dot to dot, unlike the stochastic luminescence intensity fluctuations of QD-585 due to the averaged effect of long- and short-lived "on"−"off" states. Indeed, under linearly polarized laser excitation the PL intensity varied among individual CdSe QDs, and also for the same QDs PL intensity varied with changes in the laser polarization angle. Typical PL images obtained for CdSe

QDs by exciting with linearly polarized laser at different polarization angles (0°, 45°, 90°, and 135°) are shown in Figure 3A−D. PL intensity variations of CdSe QDs with varying excitation polarization angle indicate anisotropic transition dipole orientations, characteristic of single QDs. On the other hand, PL measurements made on a group of QDs likely show negligible excitation polarization dependency due to isotropic transition dipole orientations in a group. These observations supported that subsecond PL intermittency is characteristic of single CdSe QDs. The possibility that aggregates of CdSe QDs are included in PL intensity trajectories was further ruled out by recording AFM and TEM images of QD samples from solutions of different concentrations. We prepared samples for AFM imaging from 0.05, 0.1, and 1 nM CdSe QD solutions (in 1:10 TOPO:1-butanol mixture) by spin coating on glass coverslips and mica plates. Typical AFM image of a QD sample prepared from 1 nM CdSe QD solution is shown in Figure 3E. Isolated QDs are always observed based on height measurements; an average height of 3.3 nm was identified. AFM is sensitive to height measurements, and the lateral resolution is always exaggerated due to AFM tip convolution. Therefore, we recorded and analyzed TEM images also. Typical TEM image of a sample prepared by dispersing a 1 nM CdSe QD solution on a carbon coated copper grid is shown in Figure 3F. Note that the AFM image and TEM image are obtained for samples prepared from a solution of 2 orders of magnitude higher CdSe QD concentration compared to the samples used in optical measurements. Well-separated QDs are due to TOPO capping and hydrophobic interactions between nonpolar *octyl* chains. Polarized excitation, AFM, and TEM supported aggregation of QDs under our experimental conditions was rare. Therefore the subsecond PL intermittencies are characteristic of single CdSe QDs.

We determined the PL intermittency time of individual QDs by recording and analyzing intensity trajectories at subvideo rates by exciting with a circularly polarized laser. The use of a circularly polarized laser was useful in recording and comparing trajectories of different QDs independent of their transition dipole orientations. The PL intermittency time of individual QDs was determined by autocorrelation analyses of intensity trajectories. A histogram of the intensity trajectory autocorrelation time is shown in Figure 4. An average PL intermittency time of 213 ms was obtained from autocorrelation analyses of intensity trajectories of >100 single CdSe QDs. A typical PL intensity autocorrelation decay of a single CdSe QD is shown in the inset of Figure 4. In contrast to previous power-law distribution of trajectory "off" times,[14,15] the PL intensity trajectory autocorrelations of CdSe QDs showed nearly single-exponential decays.

Luminescence intermittencies of QDs are due to charge localization and recapture. Subsecond PL intermittencies and short-lived "on"−"off" states observed in the current work are attributed to a high density distribution of homogeneous surface trap states. In previous reports, different "off" times extending over a milliseconds to minutes time scale recorded in trajectories are explained in terms of inhomogeneous distribution of trap states and disordered charge recombination.[4,11−16,18,19,25,28] In QDs with high density and varying energy surface trap states, hopping of an excited electron among trap states followed by multiple ways of charge recombination is assumed to induce time-averaged long-lived dark states, variations of blinking time, and nonexponential luminescence decays. This assumption is partly substantiated from the suppression of blinking by incorporating thiol molecules on the surface of QDs[13] and

Figure 4. Histogram of PL intermittency time constants obtained from intensity trajectory autocorrelations of >100 single CdSe QDs. Trajectories recorded at 20 ms time resolution were used in autocorrelation analyses. Inset: typical PL intensity trajectory autocorrelation trace of a single CdSe QD.
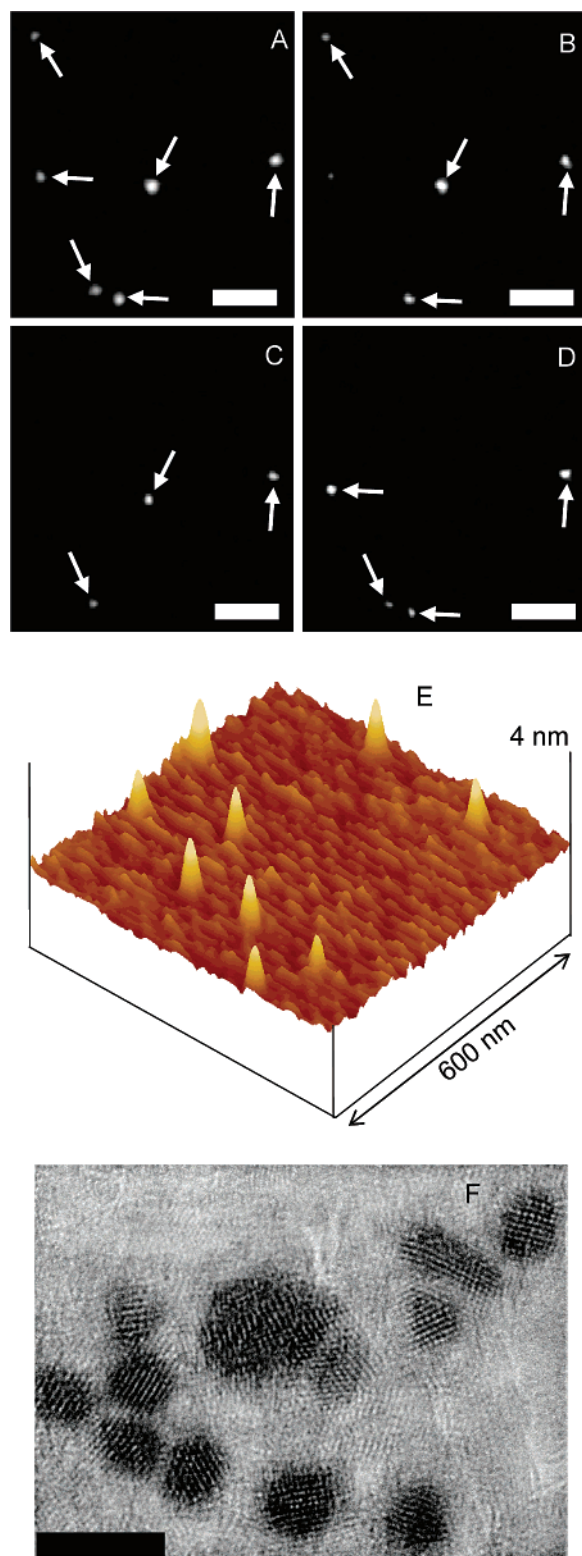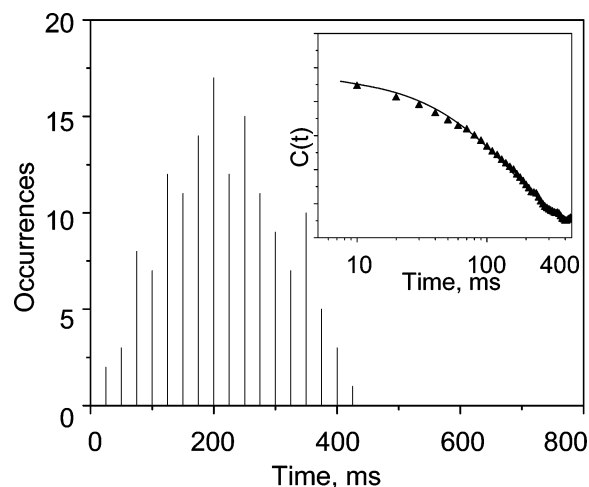

Figure 3. (A−D) PL images of single CdSe QDs obtained under linearly polarized laser excitations of different polarization angles: (A) 0°, (B) 45°, (C) 90°, and (D) 135°. Variations of PL intensity with different excitation polarizations are due to different transition dipole orientations, characteristic of single QDs. (E) AFM tapping-mode image of isolated CdSe QDs; average size is 3.3 nm from height measurements. The AFM image was collected in air for a sample prepared by spin coating a 1 nM CdSe QD solution (in 1:10 TOPO:1-butanol mixture) on a freshly cleaved mica plate. (F) TEM image of isolated CdSe QDs in a sample prepared by incubating a 1 nM CdSe QD solution (in 1:10 TOPO:1-butanol mixture) on a carbon coated copper grid. The scale bars in (A−D) are 10 μm and that in (F) is 5 nm.

distributing QDs on rough metal films,[40] identifying the contribution of thermionic emission to the lifetime and intermittency of single QDs,[11] and emission intensity dependent PL decays.[28] Furthermore, properties of QDs including luminescence color, emission bandwidth, luminescence quantum efficiency, crystal structure, and electronic defects vary considerably depending on their growth conditions such as temperature, precursor reagents, chelating medium, sequence of addition of precursor reagents, and ratio of precursors.[1,41,42,45] From previous reports on the time-averaged effect of nonexponential PL decays and intensity-dependent decays, a correlation between inhomogeneous distribution of trap states and power-law statistics of "on"−"off" time distributions is pertinent to QDs. Multiexponential PL decays identified for single QDs are attributed to dynamic fluctuations of excited-state deactivations due to the presence of different surface trap states. Considering nearly single-exponential PL intensity trajectory autocorrelation decays of CdSe QDs in the current work, we propose that homogeneous surface trap states are distributed in CdSe QDs. "Homogeneous" in the present case is an averaged effect within 20 ms trajectory resolution. By using 100 ms and 20 ms time resolution of trajectory recording, we were able to record only a portion of the intermittencies. Time-resolved fluorescence investigations and better time resolution of trajectory recording are necessary for obtaining further details of the PL intermittency.

Subsecond PL intermittency observed in the current work and dynamic fluctuation of PL in previous reports attracted us to consider general events in PL trajectories of single QDs: (a) long-lived "on" and "off" states, (b) long-lived "on" states and short-lived "off" states, (c) short-lived "on" states and long-lived "off" states, and (d) short-lived "on" and "off" states. From the present status of single QD PL investigations, a QD with essentially no trap state is expected to be nonblinking (always "on" state) on a millisecond or longer time scale. When trap states are present, excited electrons are trapped, resulting in "off" states. The above four events in PL intermittencies may be rationalized in terms of distribution of trap states and their heterogeneity (varying electron affinity or energy level): (i) a low density of trap states provides a low probability of electron trapping and long-lived "on" states; (ii) a high density of trap states provides a high probability of electron trapping and short-lived "on" states; (iii) a distribution of heterogeneous trap states can support hopping of electrons among trap states and long-

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lived "off" states; and (iv) homogeneous trap states produce short-lived "off" states. A combination of two or more of these possibilities is likely involved in the general blinking behavior of QDs with intermittencies extending on a wide time scale. In the current work we observed short-lived "on" and "off" states, possibly due to high density and homogeneity of trap states. However, the presence of trap states provided essentially no deep trap emission in the PL spectrum. This may be due to nonradiative deactivation of electrons from the trap states. Although the "off" periods are short in the present case, PL intermittency is significant due to short-lived "on" states.

Conclusions

We identified subsecond PL intensity fluctuations of CdSe QDs prepared at a slow rate by lowering the QD growth temperature. By recording and analyzing the PL intensity trajectories of single CdSe QDs, the average PL intermittency time of 213 ms was identified. On the basis of PL quantum efficiency, intensity fluctuations of single QDs, and literature reports, we attribute the subsecond intermittency of CdSe QDs to a high density distribution of homogeneous surface trap states. Rather than identifying a specific case of PL intermittency of CdSe QDs, the current work shows future possibilities for developing QDs with essentially no blinking on a longer time scale. A time-resolved spectroscopic investigation is underway in our laboratory and is expected to identify a quantitative relation between subsecond PL intermittency and excited-state deactivation of CdSe QDs.

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