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Description	

Field-induced control of universal fluorescence intermittency of a quantum dot light emitter

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With the nonstochastic quantum mechanical study of a quantum dot light emitter, we find that fluorescence intermittency statistics are universal and insensitive to the microscopic nature of the tunneling fluctuation between quantum dot and trapping state. We also investigate the power-law exponent θ and the crossover time τ_C of the *on*-time (τ_{on}) probability $P(\tau_{\mathrm{on}}) \propto \tau_{\mathrm{on}}^{-\theta}$ (for $\tau_{\mathrm{on}} \lesssim \tau_C$) and $\propto e^{-\Gamma \tau_{\mathrm{on}}}$ (for $\tau_{\mathrm{on}} \gtrsim \tau_C$) under an optical field of given energy and strength. For easy off-resonance excitation, it is found in both numerical and analytic ways that τ_C^{-1} is proportional to the intensity of the optical field (i.e., the square of the field strength) independent of the internal parameters of a quantum dot. Furthermore, it is also found that $\theta=2$ in the limit of vanishing field strength is the upper bound of the exponent and θ becomes less than 2 as the field strength increases.
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I. INTRODUCTION

The intermittency of fluorescence of single atoms or ions has been predicted as a detection method of the "quantum jump"[1] originally proposed by Bohr almost a century ago, and the first experimental observation was for the spectrum of Ba^+ ions in a radio-frequency trap.[2] To date, many molecular and nanoscopic systems, that is, colloidal semiconductor nanocrystals and quantum dots (QDs), fluorescent green proteins, organic dye molecules, and polymer nanoparticles, have been observed to have fluorescence intermittency under continuous wave (cw) irradiation.[3–6] These unique optical properties are attracting much attention because of their great potential for nanoscopic light sources that can be sustained from microseconds to many minutes. For instance, such systems have been tried as the fluorescent markers of biological processes[7] or single-photon sources for optoelectronic applications.[8] It is a big challenge to control the fluorescence of these nanoscopic building blocks to serve particular applications with proper understanding of fluorescence intermittency.

One of the most intriguing features is that the fluorescence intermittency follows power-law statistics through the *on*-time probability $P(\tau_{\mathrm{on}}) \propto \tau_{\mathrm{on}}^{-\theta}$,[3–6] where τ_{on} is the length of the "on" sojourn time and the exponent θ has been mostly found in the range between 1 and 2 peaking at about 1.5. It is surprising that the on (or "off") sojourn time is not exponentially distributed as expected from the early random telegraph model by Efros and Rosen,[9] but described by the power law implying scale invariance. Such scale invariance could be understood in terms of the dynamics of maintaining a long-memory. It was in fact reported that the process behind the subsequent on and off times of blinking is not memoriless, especially in the short time range following the power-law statistics.[10] However, it was also found that as time proceeds, the power-law statistics cross over to the bending tail of $e^{-\Gamma \tau_{\mathrm{on}}}$ near $\tau_{\mathrm{on}} \sim \tau_C$.[11] τ_C is the time scale that signifies a crossover from the long-memory to memoriless (i.e., Markov process) dynamics.

Despite its importance to both fundamental scientific profundity and technological application, the origin of QD fluorescence intermittency still remains an unsolved problem under high controversy. Since the *exponential* random telegraph model,[9] a series of modifications have been suggested.[3–6] For example, to reproduce the prevailing power-law behavior, spectral diffusion of the trapping state,[12] uniform spatial distribution of the traps,[13] spatial diffusion of the ejected electron,[14] fluctuation of nonradiative recombination,[15] a fluctuating barrier,[16] and a one-dimensional random walk[17] have been proposed. The unified picture of these models is to interpret the intermittency blinking as the charge fluctuation between a neutral (bright) and an ionic (dark) state. As for the nature and dynamics of the trapping states, however, there has been sharp disagreement among the models. Incidentally, because of the stochastic feature in common, it is rather unclear how macroscopic observations of the emission time-series would be connected to the microscopic physical quantities of QD or the details of the external optical field.

Recently, Lee and Maenosono[18] proposed a new three-state model incorporating the random fluctuation of the tunneling (or coupling) strength between a QD and the trapping state. They succeeded in achieving robust power-law behavior with an exponentially bending tail (over $\tau_{\mathrm{on}} \sim \tau_C$) through the microscopic and nonstochastic treatment of the dynamics of the light-emitting QD. The model allows the study of interacting QDs or connects the macroscopic intermittency directly to the microscopic quantities of the QD. Taking into account the Coulomb interaction between QDs could consistently explain the enhanced blinking of assembled CdSe–ZnS QDs.[19]

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In this paper, based on the recent proposition of nonstochastic quantum mechanical model of a QD light emitter,[18] we show that the fluorescence intermittency statistics are universal with little dependence on the microscopic nature of the random fluctuation of tunneling between QD and trapping state. We then investigate the central quantities of the intermittency statistics under an external optical field of energy ω and strength A in both numerical and analytic ways. For easy off-resonance condition, $\omega \neq \delta E$ (where δE is the QD excitation energy), we find that τ_C is scaled to be $\tau_C^{-1} \propto A^2$. Furthermore, we find that $\theta=2$ in the limits of $A \to 0$ and $v_0 \to 0$ (where v_0 is the strength of the tunneling fluctuation) is the upper bound of the exponent and θ becomes less than 2 as A or v_0 increases.

The paper is organized as follows. In Sec. II, we introduce a three-state model and describe the formulation. In Sec. III, through the numerical solution, we scrutinize the field-induced control of the fluorescence intermittency whose statistics are universal and insensitive to the microscopic nature of the tunneling fluctuation. In Sec. IV, through the analytic treatment, we confirm the field-induced fluorescence intermittency which is found in the numerical solution. In Secs. V and VI, the discussion and conclusion are provided, respectively.

II. MODEL AND FORMALISM

Considering the three-state model for a single QD and a trapping state, we write the Hamiltonian $\mathcal{H}$ under cw irradiation as

$$\mathcal{H} = E_\alpha c_\alpha^\dagger c_\alpha + E_\beta c_\beta^\dagger c_\beta + E_\gamma c_\gamma^\dagger c_\gamma + v(\tau)[c_\beta^\dagger c_\gamma + c_\gamma^\dagger c_\beta] + A[e^{i\omega\tau} c_\alpha^\dagger c_\beta + e^{-i\omega\tau} c_\beta^\dagger c_\alpha]. \tag{1}$$

E_α and E_β are the energies of two internal (neutral) levels of the QD and E_γ is the trapping (ionic) level.[20] $c_\alpha^\dagger(c_\alpha)$, $c_\beta^\dagger(c_\beta)$, and $c_\gamma^\dagger(c_\gamma)$ are the creation (annihilation) operators of the corresponding three states. $v(\tau)$ is the tunneling (or coupling) strength to the trapping level. The last term of $\mathcal{H}$ is the external optical pumping of energy ω and field strength A by cw irradiation.

For theoretical treatment of the photoinduced fluorescence of the QD light emitter, we directly solve the time-dependent Schrödinger equation.[21] The quantum mechanical state vector $|\psi(\tau)\rangle$ is written as

$$|\psi(\tau)\rangle = C_\alpha(\tau)e^{-iE_\alpha\tau}|\alpha\rangle + C_\beta(\tau)e^{-iE_\beta\tau}|\beta\rangle + C_\gamma(\tau)e^{-iE_\gamma\tau}|\gamma\rangle, \tag{2}$$

where $|\alpha\rangle = c_\alpha^\dagger|0\rangle$, $|\beta\rangle = c_\beta^\dagger|0\rangle$, $|\gamma\rangle = c_\gamma^\dagger|0\rangle$, and $|0\rangle$ is the vacuum. The initial state $|\Psi(0)\rangle$ should be the ground state, i.e., $|\Psi(0)\rangle = |\alpha\rangle$. The time-dependent Schrödinger equation $i\partial/\partial\tau|\Psi(\tau)\rangle = \mathcal{H}|\Psi(\tau)\rangle$ gives coupled differential equations for the coefficients $C_\alpha(\tau)$, $C_\beta(\tau)$, and $C_\gamma(\tau)$,

$$i\frac{\partial}{\partial\tau}C_\alpha(\tau) = Ae^{i\bar{\omega}\tau}C_\beta(\tau),$$

$$i\frac{\partial}{\partial\tau}C_\beta(\tau) = Ae^{-i\bar{\omega}\tau}C_\alpha(\tau) + v(\tau)C_\gamma(\tau)e^{i(E_\beta - E_\gamma)\tau}, \tag{3}$$

$$i\frac{\partial}{\partial\tau}C_\gamma(\tau) = v(\tau)C_\beta(\tau)e^{-i(E_\beta - E_\gamma)\tau},$$

where we note that the energy or time quantities are to be scaled by ω and redefine them to be dimensionless, that is, $\omega\tau \to \tau$, $E_\alpha/\omega \to E_\alpha$, $E_\beta/\omega \to E_\beta$, $E_\gamma/\omega \to E_\gamma$, $A/\omega \to A$, and $v(\tau)/\omega \to v(\tau)$. With the dimensionless parameters, $\bar{\omega}$ is defined as $\bar{\omega} = 1 - \delta E$ and $\delta E = E_\beta - E_\alpha$. Therefore, $\delta E \neq 1$ refers to the off-resonance excitation and $\delta E = 1$ the resonant excitation. It is worthy of noting that most of previous theoretical approaches are based on the classical rate equation. In those classical approaches, however, the explicit excitation processes by the external optical field might not be properly incorporated. Therefore, the present quantum mechanical approach should be consistent with the most important motivation of our study, i.e., the field-induced control of dynamics.

The microscopic randomness of the tunneling strength can be implemented in the time sequence of $v(\tau)$. Its physical origin could be discussed based on the fluctuating barrier model by Kuno and Nesbitt.[16] Further, the fluctuation would be originated from changes of the local environment. It has been argued using the Wentzel–Kramers–Brillouin theory that a $\pm 25\%$ fluctuation in the height of a 4 eV barrier between a QD and trapping state would change tunneling rates by more than a factor of 10^4, i.e., nearly the full dynamic range observed experimentally. This would naturally postulate that the tunneling strength $v(\tau)$ fluctuates almost randomly at each charge transfer. Although such a microscopic nature of $v(\tau)$ should depend on the specific system, the microscopic modeling of the time sequence of $v(\tau)$ for the system would be an improbable and formidable task. Instead, in our study, the problem is approached by assuming a few different types of random time-sequences for $v(\tau)$: (i) binary random sequence of $v(\tau)=0$ or v_0, (ii) uniform random sequence generated from $v(\tau) \in [0, v_0]$, (iii) Gaussian random sequence, or (iv) pseudorandom sequence (random sequence repeating with a finite period τ_{pseudo}). These random time sequences are illustrated in Fig. 1. v_0 could possibly be determined for a specific system.

III. NUMERICAL RESULTS

We solved the coupled differential equations of $C_\alpha(\tau)$, $C_\beta(\tau)$, and $C_\gamma(\tau)$ for $0 < \tau < 10^9$ with a time step of $d\tau = 0.01$ and a bin time of $\delta\tau = 100$. We adopt the parameters $E_\alpha = 0$, $E_\beta = 0.8$, and $E_\gamma = 1$ unless mentioned otherwise. The unnormalized probability $P(\tau_{\text{on}})$ of the on sojourn time τ_{on} is obtained by counting how many times a particular event of τ_{on} happens in the time-series of the fluorescence spectrum $\mathcal{I}(\tau)$. Also, the event of τ_{on} is defined as the on signal continues in n consecutive bin times satisfying $n = \tau_{\text{on}}/\delta\tau$. $\mathcal{I}(\tau)$ is given by $\zeta(\tau)\Theta(\zeta(\tau))$ with $\zeta(\tau) = |C_\alpha(\tau)|^2 + |C_\beta(\tau)|^2 - 0.65$,[22] where $\Theta(x)$ is the Heaviside step function. In Fig. 2, the

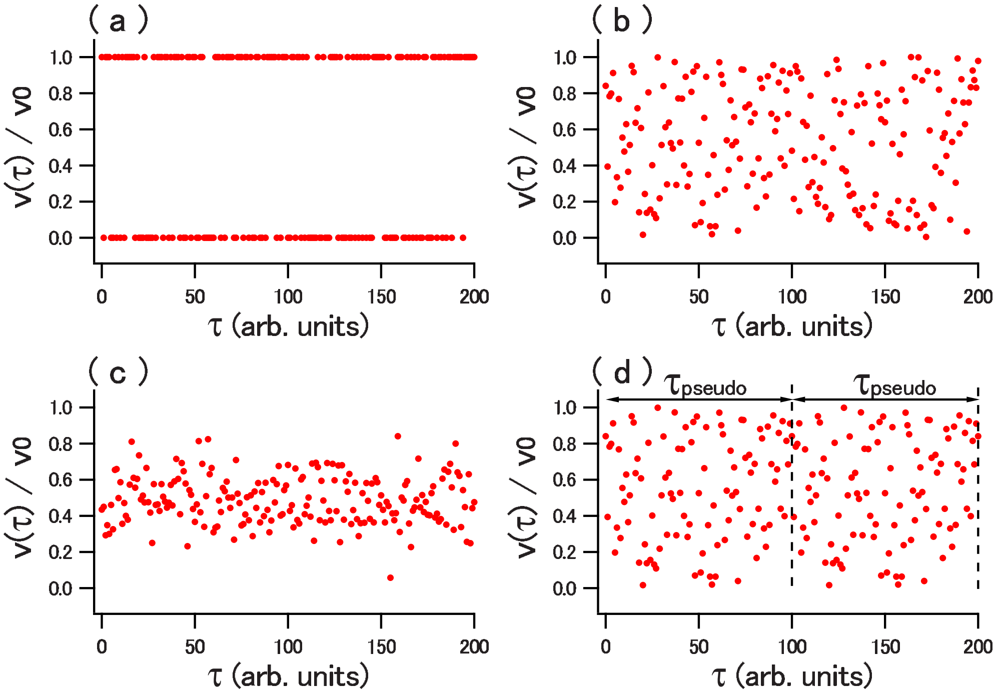

FIG. 1. Illustration of sample random sequences for $v(\tau)$: (a) binary random sequence of 0 or v_0, (b) uniform random sequence of $v(\tau)\in[0,v_0]$, (c) Gaussian random sequence with an average value $\bar{v}(=v_0/2)$ and variance $\sigma(=0.2v_0)$, and (d) pseudouniform random sequence repeating with a period τ_{pseudo}.

statistics of $P(\tau_{\text{on}})$ are provided with respect to the strength A of the external optical field for the binary random sequence (say type I) and the uniform random sequence (say type II) for $v(\tau)$. For both types, the inverse crossover time τ_C^{-1} is found to have quadratic dependence on A and θ is found to decrease as A increases. This corresponds to $\tau_C^{-1}\propto\mathcal{P}$, where $\mathcal{P}$ is the laser intensity ($\propto A^2$). Experiments of the linear laser-intensity-dependence have been reported by Stefani *et al.*[23] (for $Zn_{0.42}Cd_{0.58}Se$ QD) and Lee *et al.*[24] (for CdSe/ZnS QD) for both on the insulating glass, consistent with an isolated QD emitter in our case. The decreasing behavior of θ with A has also been observed recently for CdSe/ZnS QDs by Peterson and Nesbitt.[25] In the left panel of Fig. 3, for types I and II sequences for $v(\tau)$, the behaviors of τ_C are displayed with respect to the inverse tunneling strength $1/v_0$. It is found that τ_C is scaled to be $\tau_C\propto 1/v_0$ except for $v_0 \gg 1$ for both sequences of $v(\tau)$. The dependences of τ_C on A or v_0 could also give the important insight on the suitable parameter set necessary to compare with the actual experiment. Here we mention that the statistics of the off sojourn time τ_{off} is symmetric to those of τ_{on} in our study because of neglect of relaxation channels.

Figures 2 and 3 (left panel) imply that the fluorescence intermittency statistics have little dependence on the microscopic nature of the tunneling fluctuation. One may predict the QD dynamics under other random sequences for $v(\tau)$. For the Gaussian random sequence, $\sigma\gg 1$ leads to a situation qualitatively similar to that of type II sequence, while $\sigma\ll 1$ [i.e., $v(\tau)\to\bar{v}$] does not result in fluorescence intermittency, but steady fluorescence. For a pseudorandom sequence, i.e., a random sequence repeating with a finite period τ_{pseudo}, it is noted that the QD dynamics and fluorescence intermittency are governed by the competition between the binning time $\delta\tau$ and τ_{pseudo}. We find that the power-law intermittency is reproduced for $\tau_{\text{pseudo}}\gg\delta\tau$, but not for $\tau_{\text{pseudo}}\lesssim\delta\tau$, as illustrated in Fig. 4.

IV. ANALYTIC RESULTS

In this section, we seek a deeper physical insight through the analytic treatment of the same problem. Our model is simple enough to enable some limited analytic arguments. Starting from $v(\tau)=0$, we can obtain the exact solutions as

$$C_\alpha^{(0)}(\tau)=e^{i(\bar{\omega}/2)\tau}\left[\frac{A^2}{\bar{\omega}X+2X^2}e^{iX\tau}-\frac{A^2}{\bar{\omega}X-2X^2}e^{-iX\tau}\right],$$

$$C_\beta^{(0)}(\tau)=-i\frac{A}{X}e^{-i(\bar{\omega}/2)\tau}\sin X\tau, \tag{4}$$

where $X=\sqrt{A^2+\bar{\omega}^2/4}$. Of course, we have $C_\gamma^{(0)}(\tau)=0$. Using the exact solutions of Eq. (4), $C_\gamma^{(1)}(\tau)$ can be immediately obtained from Eq. (3) as

$$C_\gamma^{(1)}(\tau)=-\frac{A}{X}\int_0^\tau d\tau' v(\tau')\sin X\tau' e^{-i(\bar{\omega}/2)\tau'}e^{-i(E_\beta-E_\alpha)\tau'}. \tag{5}$$

$C_\gamma^{(3)}(\tau)$ can also be easily obtained after getting $C_\beta^{(2)}(\tau)$ by applying the successive perturbation scheme to Eq. (3),

$$C_\gamma^{(3)}(\tau)=\frac{A}{X}\int_0^\tau d\tau' v(\tau')e^{-i(E_\beta-E_\gamma)\tau'}$$
$$\times\int_0^{\tau'} d\tau'' v(\tau'')e^{i(E_\beta-E_\gamma)\tau''}$$
$$\times\int_0^{\tau''} d\tau''' v(\tau''')\sin X\tau''' e^{-i(\bar{\omega}/2)\tau'''}e^{i(E_\beta-E_\gamma)\tau'''}. \tag{6}$$

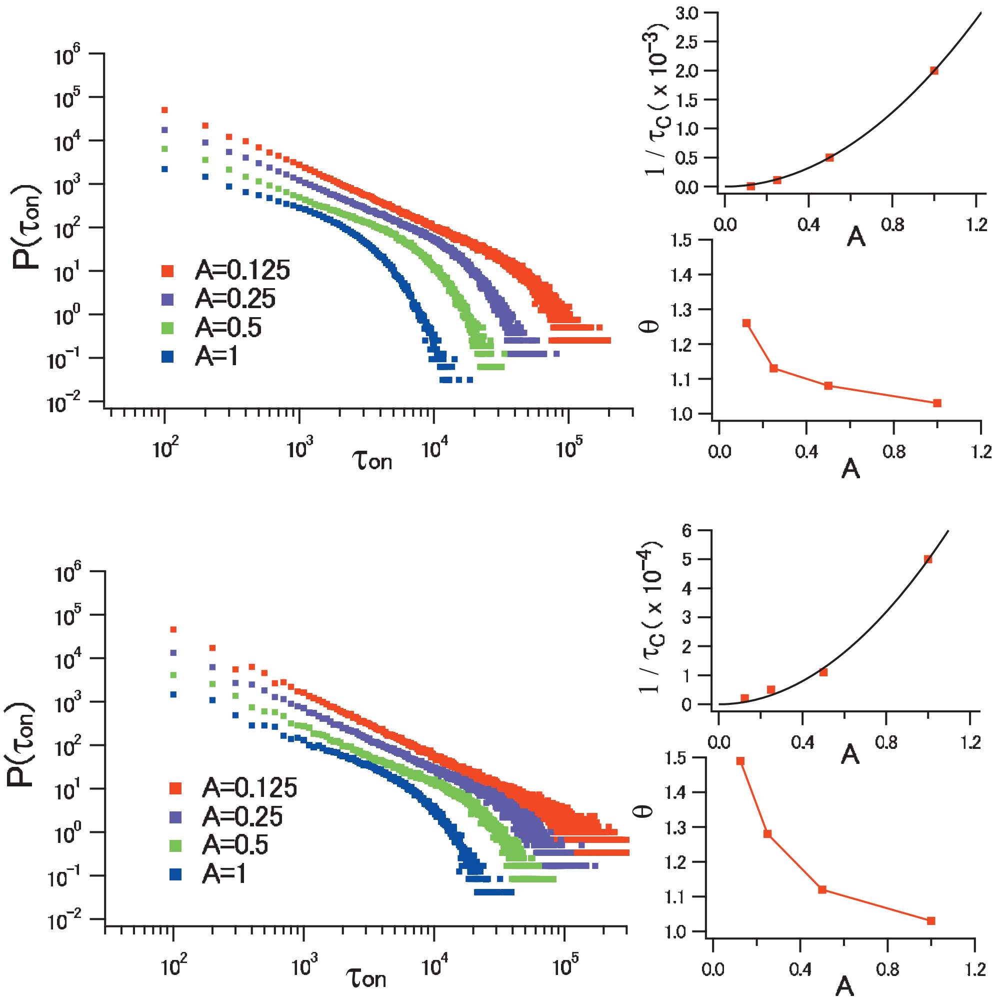

FIG. 2. Unnormalized probability of the on sojourn time for types I (upper panel) and II (lower panel) sequences of $v(\tau)$ with respect to A: For both types, $v_0=1$ is used. Behaviors of $1/\tau_C$ and θ are also given with respect to A.

Both Eqs. (5) and (6) can be examined in the following interesting limits. First, we assume off-resonance excitation, i.e., $\bar{\omega}\neq 0$ or $\delta E\neq 1$. This is a straightforward situation because the realistic electronic structure of a QD comprises multiple conduction and valence levels. Second, we assume $E_\beta \approx E_\gamma$. As shown in the right panel of Fig. 3, the relative position of E_β and E_γ is not so relevant. This is interesting because it may imply that the spectral diffusion of trapping states[12] might not be consistent with the blinking statistics. Third, we examine the equations in the limits of the weak field strength (i.e., $A\rightarrow 0$) and the weak tunneling strength (i.e., $v_0\rightarrow 0$). Under these considerations, picking

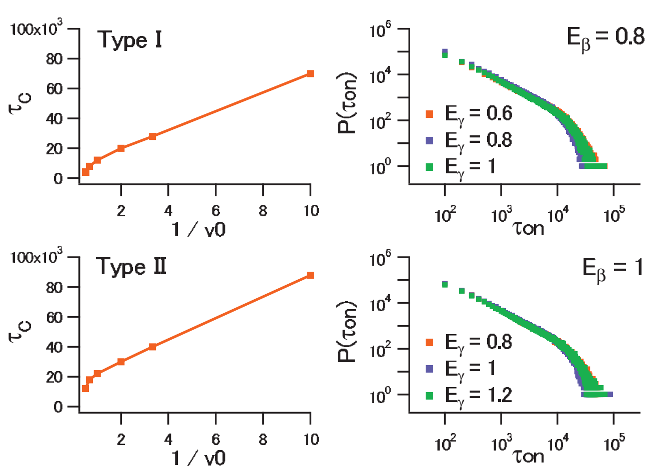

FIG. 3. Left panel: Behavior of τ_C with respect to $1/v_0$ for types I and II sequences of $v(\tau)$. $A=0.25$ is used. Right panel: Unnormalized probability of the on sojourn time with E_γ and E_β ($E_\alpha=0$). $E_\beta=0.8$ and $E_\beta=1$ correspond to off-resonance and resonant excitation, respectively. Type I sequence for $v(\tau)$ with $v_0=1$ and $A=0.25$ is used.

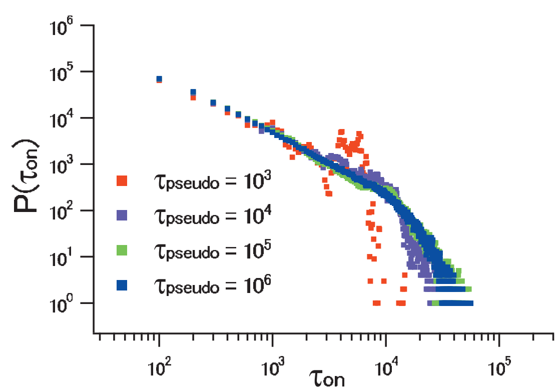

FIG. 4. Unnormalized probability of the on sojourn time for the bin time $\delta\tau=100$ with several values of τ_{pseudo}: The pseudobinary random sequence (repeating with a period τ_{pseudo}) with $v_0=1$ and $A=0.25$ are used.

up only the slowly oscillating contribution with $X\approx\bar{\omega}/2+A^2/\bar{\omega}+\cdots$, we find

$$C_\gamma^{(1)}(\tau)\approx i\frac{A}{2X}\int_0^\tau d\tau' v(\tau')e^{-i(A^2/\bar{\omega})\tau'}$$
$$\approx i\eta_1 v_0 A\tau+\eta_2 v_0 A^3\tau^2+\cdots \quad (7)$$

and in the same way,

$$C_\gamma^{(3)}(\tau)\approx i\eta_3 v_0^3 A\tau^3+\cdots, \quad (8)$$

where η_1, η_2, and η_3 are real numbers of $\mathcal{O}(1)$ with bounded randomness, such as $\eta\in[\bar{\eta}-\delta\eta/2,\bar{\eta}+\delta\eta/2]$. $\bar{\eta}$ is an average value.

Now, let us discuss the intermittency statistics on the basis of the above analytic exploration. We define the probability that the state is unchanged during the time interval $\tau_0\le\tau\le\tau_0+\mathcal{T}$ when the system is in a bright (neutral) state initially at τ_0 as $P(\tau_0,\tau_0+\mathcal{T})$. Then $P(\tau_0,\tau_0+\mathcal{T})$ can be formally written as

$$P(\tau_0,\tau_0+\mathcal{T})=P(\tau_0,\tau_0+\delta\tau)P(\tau_0+\delta\tau,\tau_0+2\delta\tau)$$
$$\times\cdots\times P(\tau_0+(N-1)\delta\tau,\tau_0+\mathcal{T}), \quad (9)$$

where $\mathcal{T}=N\delta\tau$. Here we note an expression of $P(\tau_0,\tau_0+\delta\tau)$ in terms of $|C_\gamma(\tau)|^2$ simply like

$$P(\tau_0,\tau_0+\delta\tau)\approx\frac{|C_\gamma(\tau_0)|^2}{|C_\gamma(\tau_0+\delta\tau)|^2}. \quad (10)$$

With $|C_\gamma(\tau)|^2=|C_\gamma^{(1)}(\tau)+C_\gamma^{(3)}(\tau)+\cdots|^2$, we obtain the first few leading order terms for $P(\tau_0,\tau_0+\mathcal{T})$ in the limits of $A\to 0$ and $v_0\to 0$,

$$P(\tau_0,\tau_0+\mathcal{T})\propto\frac{1}{\eta_1^2\mathcal{T}^2+6\tau_0\left(\eta_1\eta_3 v_0^2+\frac{2\eta_2^2}{3}A^4\right)\mathcal{T}^3+\cdots}$$
$$\approx\frac{1}{\eta_1^2}\mathcal{T}^{-2}-6\tau_0\left(\frac{\eta_3}{\eta_1^3}v_0^2+\frac{2\eta_2^2}{3\eta_1^4}A^4\right)\mathcal{T}^{-1}+\cdots$$
$$\propto\mathcal{T}^{-2}+(\nu-\Gamma)\mathcal{T}^{-1}+\cdots, \quad (11)$$

where we assume $\mathcal{T}\gg\tau_0$ keeping $\tau_0\mathcal{T}\sim\mathcal{O}(1)$ (τ_0 is arbitrary). The last line of Eq. (11) is for a comparison with an empirical formula $P(\tau_0,\tau_0+\mathcal{T})\propto\mathcal{T}^{-\theta}e^{-\Gamma\mathcal{T}}$, with $\theta=2-\nu$. Noting that Γ is known to be $\Gamma\propto\tau_C^{-1}\propto[\chi v_0+\zeta A^2]$ from the behaviors of τ_C shown in Figs. 2 and 3, we finally find $\nu\propto[\chi v_0+\zeta A^2+\mathcal{O}(v_0^2,A^4)]$. Therefore, Eq. (11) shows that the power-law exponent has $\theta=2$ as its upper bound in the limits of $A\to 0$ and $v_0\to 0$ and becomes less than 2 as A or v_0 increases. The sketch of the behavior of θ with respect to A is displayed in Fig. 5. This naturally explains the decrease of θ with increasing A at the qualitative level, already shown in Fig. 2.

The power-law behavior changes to the exponential behavior near the crossover time τ_C. Such a crossover means a change from the long-memory to memoriless dynamics. The lowest order term of $C_\gamma(\tau)$ thus has the randomness of $\delta\eta_1 v_0 A\tau$ centered about $\bar{\eta}_1 v_0 A\tau$ and in the same way, the next higher order terms have randomness of $\delta\eta_2 v_0 A^3\tau^2$ and $\delta\eta_3 v_0^3 A\tau^3$. If the randomness of the next higher order term is larger than that of the lowest order term, the initial memory

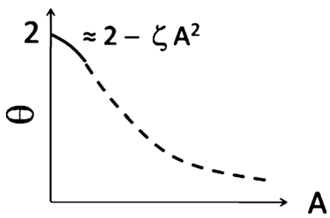

FIG. 5. Sketch of the power-law exponent θ with respect to the field strength A (for $v_0\to 0$): The dashed part is from the numerical results in Fig. 2.

disappears and the dynamics cross over to the memoriless ones. That is, we impose for the crossover time τ_C,

$$v_0 A\tau_C\sim v_0 A^3\tau_C^2 \quad\text{and}\quad v_0 A\tau_C\sim v_0^3 A\tau_C^3.$$

Therefore, we find $\tau_C^{-1}\propto A^2$ and $\tau_C^{-1}\propto v_0$. These relations confirm our numerical findings in Figs. 2 and 3 (left panel), respectively. We note that in the lowest order, A and v_0 do not couple to each other in the scaling of τ_C^{-1}. This signifies that the field-dependence of τ_C^{-1} is independent of the internal parameters of the QD (i.e., size of v_0 or internal levels according to Fig. 3). That is, numerical results of Fig. 2 would be robust to the value of v_0 or relative position of E_β and E_γ.

After the crossover time τ_C, the memory of the dynamics is completely lost so that one has $P(\tau_0,\tau_0+\delta\tau)=P(\tau_0+\delta\tau,\tau_0+2\delta\tau)=\cdots=C$. From this, $P(\tau_0,\tau_0+\mathcal{T})$ can be written as

$$P(\tau_0,\tau_0+\mathcal{T})=C^N=e^{-\xi(\mathcal{T}/\delta\tau)} \quad (12)$$

taking $C=e^{-\xi}$. It is readily understood that the intermittency statistics proceed from the power-law behavior to the exponential behavior.

So far, we have considered only the easy off-resonance excitation ($\bar{\omega}\neq 0$ or $\delta E\neq 1$); however, one may need to consider the resonant excitation of $\bar{\omega}\approx 0$, i.e., $\delta E\approx 1$. If a particular excitation, for instance, the lowest excitation of $1S_h\to 1S_e$ in CdSe QD, has a dominantly strong oscillator strength, one may think of the resonant excitation by tuning the field energy to the corresponding excitation energy. In this case, we have $C_\gamma^{(1)}(\tau)\approx-\int_0^\tau d\tau' v(\tau')\sin A\tau'\approx\eta_1 v_0 A\tau^2+\eta_2 v_0 A^3\tau^4+\cdots$ and in the same way, $C_\gamma^{(3)}(\tau)\approx\eta_3 v_0^3 A\tau^4+\cdots$, where η_1, η_2, and η_3 carry similar randomness. We estimate τ_C as $v_0 A\tau_C^2\sim v_0 A^3\tau_C^4$ and $v_0 A\tau_C^2\sim v_0^3 A\tau_C^4$ and find $\tau_C^{-1}\propto A$ and $\tau_C^{-1}\propto v_0$. In particular, $\tau_C^{-1}\propto A$ sharply contrasts with the relation of $\tau_C^{-1}\propto A^2$ for the easy off-resonance excitation. In Fig. 6, we confirm $\tau_C^{-1}\propto A$ through the numerical calculation.

V. DISCUSSION

The first QD blinking model was suggested by Efros and Rosen.[9] Even if the biexciton and its successive Auger ionization based on the model cannot directly predict the power-law intermittency, the physical insight by the Auger-like nonradiative relaxation is not small. We argue that such relaxation could be in principle included in our formalism and considered to contribute *effectively and partly* to the effective tunneling constant. However, it is assumed that the contribution would be eventually screened in the strong fluctuation of the direct tunneling in our consideration. This is

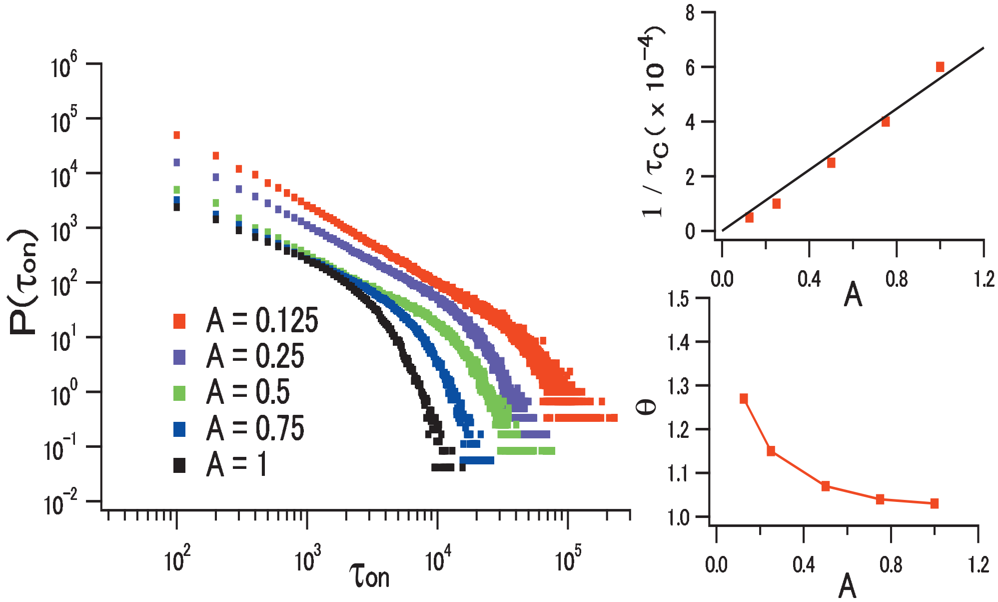

FIG. 6. Unnormalized probability of the on sojourn time with respect to A under resonant excitation, i.e., $\delta E = E_\beta - E_\alpha = 1$ ($E_\alpha = 0$, $E_\beta = 1$, and $E_\gamma = 1.2$ are taken). Type I sequence of $v(\tau)$ with $v_0 = 1$ is used. Behaviors of $1/\tau_C$ and θ are also given with respect to A.

schematically illustrated in Fig. 7. From the figure, one may note that $v + G\bar{G}v + \cdots$ would correspond to $v(\tau)$ in Eq. (1), where G and $\bar{G}$ stand for electron-hole creation and recombination, respectively. $G\bar{G}v$ would simply represent the Auger process together with the Auger electron to the trapping level.

Another point to be considered in the underlying dynamics of QD would be the radiative decay, that is, the spontaneous emission. The spontaneous emission cannot be immediately incorporated in our model Hamiltonian because it has no classical analog and should be handled in the quantum mechanical photon field. In our model, the optical pumping is done by the classical radiation field. Nevertheless, we could try to set in the relevant term by hand in the equation. In that case, we find that our finding of the field-induced scaling of the QD blinking statistics is robust because the energy scale relevant for the spontaneous emission [typically, $\sim 10^{-6}$ eV corresponding to $\sim \mathcal{O}(1)$ ns] is much smaller than the field parameters. For a given set of the QD parameters (all the parameters are scaled by ω) with $\omega = \mathcal{O}(1)$ eV, it may be possible to make an estimation for τ_C, that is, by taking $A \sim 0.1-0.01$ meV and $v_0 \sim 10$ meV, one may get $\tau_C \sim 0.1-10$ s, which is more or less comparable with the experiment. In spite of such an estimation, in order to discuss the absolute time scales, we may have limitations in our conclusion and need to explicitly consider the spontaneous emission more carefully.

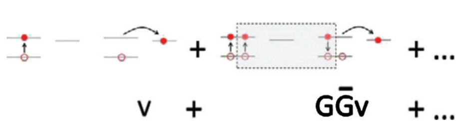

FIG. 7. Schematic illustration of the direct tunneling (v) and the biexciton and the Auger-like nonradiative relaxation ($G\bar{G}v$): The shaded box corresponds to $G\bar{G}$, where G and $\bar{G}$ represent the propagator of electron-hole creation and recombination, respectively. Filled red dots represent electrons and empty dots holes, respectively.

Finally, we remark on the field-dependence of the QD fluorescence intermittency, which might be seminal to both application control and fundamental understanding of the phenomena. With regard to the laser-intensity-dependence of the inverse crossover time τ_C^{-1}, one may find special interest because it could be directly observed in the experiments. Stefani *et al.*[23] and Lee *et al.*[24] discussed the important role of the QD's environment and reported the linear dependence for QDs on the insulating glass. In this environment, QDs would respond like isolated ones with the least communication between them, consistent with an isolated QD emitter in our present consideration. Peterson and Nesbitt[25] reported the quadratic dependence for QDs on the polymer film and argued it as evidence of the increasing probability of biexciton formation leading to Auger ionization. However, it is well known that the random telegraph model[9] which originally proposed the scenario of biexciton and Auger ionization cannot reproduce the power-law distribution of on or off times.

VI. CONCLUSION

In conclusion, we found that the intermittency statistics are universal (i.e., power-law behavior with an exponential tail) with little dependence on the microscopic nature of tunneling fluctuation between QD and trapping state. The power-law exponent θ, crossover time τ_C, and their optical field dependence were investigated in both numerical and analytic ways. We found that for easy off-resonance excitation, τ_C is scaled to be $\tau_C^{-1} \propto A^2$ ($\propto \mathcal{P}$) independent of internal parameters of QD. We also found that $\theta = 2$ in the limits of $A \to 0$ and $v_0 \to 0$ is the upper bound of the exponent and θ becomes less than 2 as A or v_0 increases.

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[1] R. J. Cook and H. J. Kimble, Phys. Rev. Lett. **54**, 1023 (1985).
[2] W. Nagourney, J. Standberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).
[3] F. Cichos, C. Von Borczyskowski, and M. Orrit, Curr. Opin. Colloid Interface Sci. **12**, 272 (2007).
[4] P. Frantsuzov, M. Kuno, B. Janko, and R. A. Marcus, Nat. Phys. **4**, 519 (2008).
[5] F. D. Stefani, J. P. Hoogenboom, and E. Barkai, Phys. Today **62**(2), 34 (2009).
[6] S. F. Lee and M. A. Osborne, ChemPhysChem **10**, 2174 (2009).
[7] M. Dahan, S. Levi, C. Luccardini, P. Rostang, B. Riveau, and A. Triller, Science **302**, 442 (2003).
[8] P. Micher, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature (London) **406**, 968 (2000).
[9] A. L. Efros and M. Rosen, Phys. Rev. Lett. **78**, 1110 (1997).
[10] F. D. Stefani, X. Zhong, W. Knoll, M. Han, and M. Kreiter, New J. Phys. **7**, 197 (2005).
[11] S. Wang, C. Querner, T. Emmons, M. Drndic, and C. H. Crouch, J. Phys. Chem. B **110**, 23221 (2006).
[12] J. Tang and R. A. Marcus, Phys. Rev. Lett. **95**, 107401 (2005).
[13] R. Verberk, A. M. van Oijen, and M. Orrit, Phys. Rev. B **66**, 233202 (2002).
[14] G. Margolin, V. Protasenko, M. Kuno, and E. Barkai, Adv. Chem. Phys. **133**, 327 (2006).
[15] P. A. Frantsuzov and R. A. Marcus, Phys. Rev. B **72**, 155321 (2005).
[16] M. Kuno, D. P. Fromm, H. F. Hermann, A. Gallagher, and D. J. Nesbitt, J. Chem. Phys. **115**, 1028 (2001).
[17] K. T. Shimizu, R. G. Neuhauser, C. A. Leatherdale, S. A. Empedocles, W. K. Woo, and M. G. Barwendi, Phys. Rev. B **63**, 205316 (2001).
[18] J. D. Lee and S. Maenosono, Phys. Rev. B **80**, 205327 (2009).
[19] M. Yu and A. Van Orden, Phys. Rev. Lett. **97**, 237402 (2006).
[20] The model is so general that another interpretation might be possible, for example, $|\beta\rangle$ and $|\gamma\rangle$ might be considered as bright and dark exciton, respectively.
[21] J. D. Lee, J. Inoue, and M. Hase, Phys. Rev. Lett. **97**, 157405 (2006); J. D. Lee and M. Hase, *ibid.* **101**, 235501 (2008).
[22] The on signal is determined by $|C_\alpha(\tau)|^2+|C_\beta(\tau)|^2 \geqslant |C_\gamma(\tau)|^2$, i.e., $|C_\alpha(\tau)|^2+|C_\beta(\tau)|^2 \geqslant 0.5$ from $|C_\alpha(\tau)|^2+|C_\beta(\tau)|^2+|C_\gamma(\tau)|^2=1$. A practical criterion is chosen to be $|C_\alpha(\tau)|^2+|C_\beta(\tau)|^2>0.65$.
[23] F. D. Stefani, W. Knoll, M. Kreiter, X. Zhong, and M. Y. Han, Phys. Rev. B **72**, 125304 (2005).
[24] D. H. Lee, C. T. Yuan, M. Tachiya, and J. Tang, Appl. Phys. Lett. **95**, 163101 (2009).
[25] J. J. Peterson and D. J. Nesbitt, Nano Lett. **9**, 338 (2009).