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NUMERICAL SIMULATION OF BLINKING QUANTUM DOT BASED ON TEMPERATURE DEPENDENCE 3-DIMENSIONAL LEVY RANDOM WALK

Anita Sharma, M.N. Bapat

Regional Institute of Education,
Bhopal, Madhya Pradesh, India
E-mail: annu5.sharma@gmail.com

Blinking statistics of quantum dot has attracted much attraction in recent years. Various experiments were conducted and various theories have been given to explain this phenomenon. However, the problem is not yet resolved. The weak temperature dependence of the power law parameters have complicated the phenomena. We have simulated the blinking statistics of quantum dot based on the random walk model. We have shown that three-dimensional biased Levy random walk of electrons, the bias being the Columbic interaction between electrons and ionized atoms can explain the observed experimental results. We have simulated the blinking properties of quantum dots in a broad temperature range (10-300K). *The distributions exhibit power law behavior for a wide range of temperature, but the power law parameter increases marginally with temperature. The trend of change is independent of the size of the quantum dots as confirmed from the simulation.*

Keywords*: QUANTUM DOT, LEVY STATISTICS, RANDOM WALK, FLUORESCENCE.*

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1. INTRODUCTION

Quantum dot (QD) devices have attracted much attraction since last decades due to interesting physical phenomena and practical applications. One of the physical phenomena that needs attention is their blinking properties under continuous excitation [1, 3]. This means while using them as fluorescent devices, they are not "switch on" all the time even though the incident excitation is continuous [4]. In dramatic contrast to the usual expectation, distributions of on and off times of QDs follow a universal power-law behavior, not the characteristic, exponential behavior of Poissonian kinetics [5], i.e. the "on time" and "off time" distributions exhibit power law as in relation (1):

$$\begin{aligned} P_{on}(t_{on}) &\propto 1/t_{on}^{m_{on}}, \\ P_{off}(t_{off}) &\propto 1/t_{off}^{m_{off}}, \end{aligned} \tag{1}$$

where t_{on} (or t_{off}) is on (or off) times and m_{on} (or m_{off}) is the power law parameter governing the on (or off) time distribution. Since a single quantum dot comprising thousands atoms is not single absorber and more than one e-h pair can be created in such QD simultaneously, it is surprising to observe their blinking photoluminescence.

Various models have been proposed to explain this phenomenon. This includes Static trap model [6, 7], Dynamic trap model [8] and Random walk model [9, 10]. In this work, we have simulated the blinking statistics of quantum dots by using random walk model. We have estimated the power law parameter for on and off time distributions and their temperature dependence (10-300 K). Similar calculations were repeated many times to evaluate the error involved in the calculation.

2. RANDOM WALK MODEL FOR QD BLINKING:

The random walk model proposed by Margolin et al. [11] gives an explanation in which the blinking by three-dimensional hopping diffusion of the photo-ejected electron is in the surrounding media (Fig. 1a). The ejected electrons roam around the QD randomly and return to the QD after many scattering event. The recombination occur in the QD, which can be radiative or non radiative. The non-radiative transition is mainly governed by the Auger transition. Auger effect is possible in the presence of an extra hole or electrons, which can take the energy (Fig. 1b). Thus Auger transition is favored for charged QD and there will be non radiative transition and is called "Auger quenching". This Auger quenching mechanism is considerably faster than the mechanism leading to luminescence. Thus, the QD is in the off state and will remain in the off state until the hole in the valence band is filled again [12]. This might occur when the diffusing electron recombines with this hole in the core. Thus the positively charged QD stays "off" until the electron returns. The long "on" times are explained by the existence of a long-lived hole trap in the vicinity of the QD. The hole is trapped to the surface state by the Auger mechanism. While the hole is trapped and the electron is diffusing, the QD stays "on." In random walk model, it was assumed that once the electrons are emitted from the QD, they undergo a random walk around the QD before recombining again.

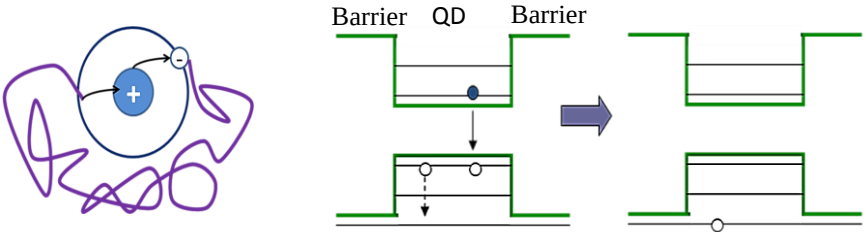

Fig. 1 *– A schematic figure of random walk model. Circles with – and + sign are electrons and positive ions respectively. The arrows indicate path of electron* (*a*). *Schematic of the Auger process in QD with respective band diagram* (*b*). *The dark circle and open circles represent electron and holes respectively*

This gives rise to blinking behavior. It can be shown numerically that if the path length of this random walk is distributed according to Levy distribution, the t_{on} and t_{off} shows power law behavior. Frequently it is cited in literature that because of the nonzero escape probability of the ejected electron from the quantum dot, this model cannot explain the observed result successfully. This is motivated by the fact that given infinite time, a random walker in 1 and 2 dimensions will return to its original place with

certainty. However in more than 2 dimensions (2D), the probability is less than one and the escape probability become non zero.

Here we simulate the random walk in 3D and restrict the electron to escape by considering the coulomb interaction between electron and charged atom. Recently we have explained successfully the power law behavior of QD blinking by random walk model [13]. To understand thoroughly the power-law behavior, in this work, we have simulated the temperature dependence of the linking statistics.

3. METHODOLOGY

We have simulated the random walk by letting the electrons move randomly in 3D in the vicinity of QD and tracking individual electrons. We have assumed that the ejection of an electron from a QD to the surrounding medium can be thermally induced or occur because of a direct photon impact. By the latter here we mean the process of a complete energy transfer from an incident photon to an electron. The barrier height was assumed to be 0.2 eV. Thus any electron in the conduction band with energy higher than 0.2 eV undergoes random walk in the vicinity of QD. The electron energy distribution in the conduction band was calculated from the incident photon energy and band gap of the QD material and was allowed to spread as Gaussian distribution with standard deviation equal to the thermal broadening at that temperature (k_BT). The effect of temperature was simulated by the thermal escape probability, Auger recombination probability and electron velocity distribution. The probability of auger process is temperature dependent and can be understood by the following argument. The initial energy of the photo-excited electrons is decided by the energy of the incident photon. However, as the electrons scatters around the QD, they lose energy and after many scattering events, their energy distribution is given by the Fermi-Dirac distribution at the temperature of the lattice. Therefore, the energy of the electrons coming back to the QD is decided by the temperature under consideration and not by the incoming light energy. The Auger recombination cross section depends on the energy of the carrier as E^3 and thus the non-radiative transition probability is highly temperature dependent [14]. In addition to the temperature dependence of Auger coefficient, the thermal ionization of the carrier in the quantum dots are also considered. We have neglected any local heating due to the incident light intensity. We define the on time (or off time) as the interval of time when no signal falls below (or surpasses) a chosen threshold intensity value. Each step is independent of the previous step. We have simulated the random walk in continuous space. So, a particle was located at the center of a three dimensional Cartesian coordinate. In each time step, the particle jumped to a location which was on the surface of a sphere with radius r, where r was a continuous Levy variable. Coulomb interaction between the ionized QD and electrons were simulated by applying bias to the random walk simulation. The bias was the ratio of electrostatic potential energy and kinetic energy of the electrons. All the simulations were done using MATLAB, which has periodicity 2^{1492} for the generation of random number, therefore the simulations can be considered reliable. Numbers of atoms inside a QD was taken as 400. Since in practice, the average QD density obtained is around 10^{10} cm^{-2}, the average distance between two QD will be 10^{-5} cm. Therefore, the maximum step length was taken to be 0.1 μm. In this calculation we have taken InAs/GaAs QD with size 5 nm, which has estimated 500 atoms.

4. RESULTS AND DISCUSSIONS

Figure 2 is the intensity distribution w.r.t time of fluorescent spectra of single QD at room temperature (300K). It clearly shows the bright and dark state of blinking QD. The respective times t_{on} and t_{off} are mentioned in the zoomed figure, where "on" state and "off" states are marked.

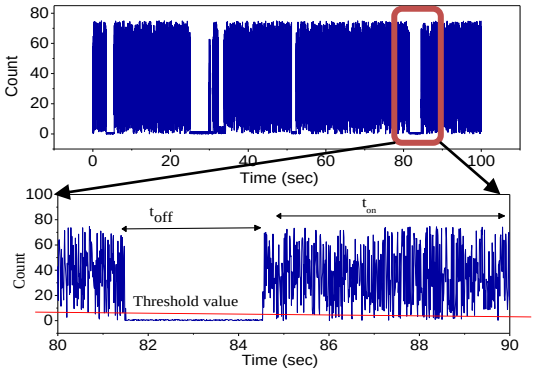

Fig. 2 *– Sample 100 sec intensity distribution of blinking QD and expanded version of the same showing on and off time*

Figures (3a) and (3b) show the temperature dependence of power law spectra at four representative temperatures (300 K, 200 K, 50 K and 10 K). As seen from the figure, both the on and off time distribution follows power law type behavior regardless of the temperature. A power law plot is seen as a straight line in double log plot and the power law parameters (m_{on} and m_{off}) is determined from the slope of the curve. From the first view at these figures, it appears that there is no significant effect of temperature on the power law spectra. However, the estimated parameters present clearer picture. The estimated values are plotted in figure 4a as a function of temperature. It can be seen that, both m_{on} and m_{off} increases with increase in temperature with noticeable linear behavior. The error bars are a result of repeated calculations. At all temperatures m_{on} is greater than m_{off}. Room temperature values of m_{on} and m_{off} match with the experimentally observed values of F. Cichos, C [12]. We could not find any experimental or theoretical report dealing with the temperature dependence of m_{on} and m_{off} over a broad range of temperature.

Another important observation in this simulation is that at higher time scale, both the on and off time distribution do not obey the exact power law, rather they obey more Gaussian like behavior. This is observed as the deviation from the linearity in the log-log curve. This type of behavior was observed by many authors [3, 6, 15] but the observation of Fernando Stefani et.al [5] on the contrary, shows power law behavior in all time scale. Since our simulation is for a biased random walk, it could be assumed that the columbic interaction between the electron and ionized QD decide the long time scale behavior. When the temperature is lowered, the average kinetic energy of the electrons reduces. Thus the bias becomes more effective. Therefore, the deviation from linearity is expected to be more significant.

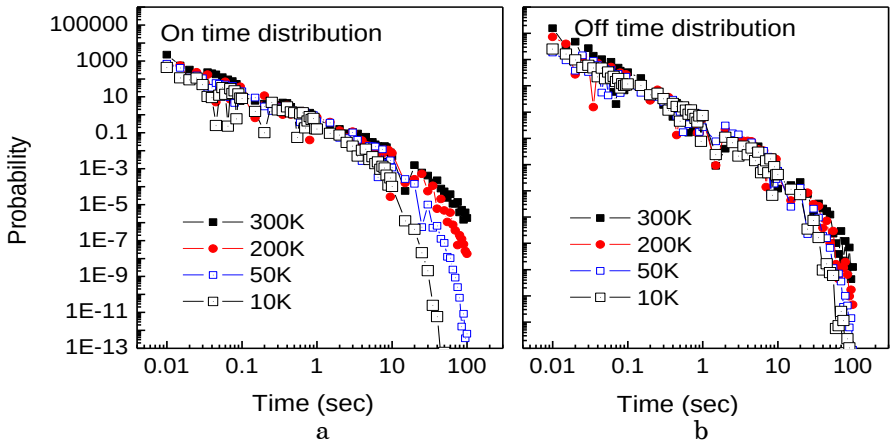

Fig. 3 – *On* (*a*) *and Off* (*b*) *time distribution of fluorescence of single QD with* 200 *atoms for different temperatures* (300, 200, 50 *and* 10K)

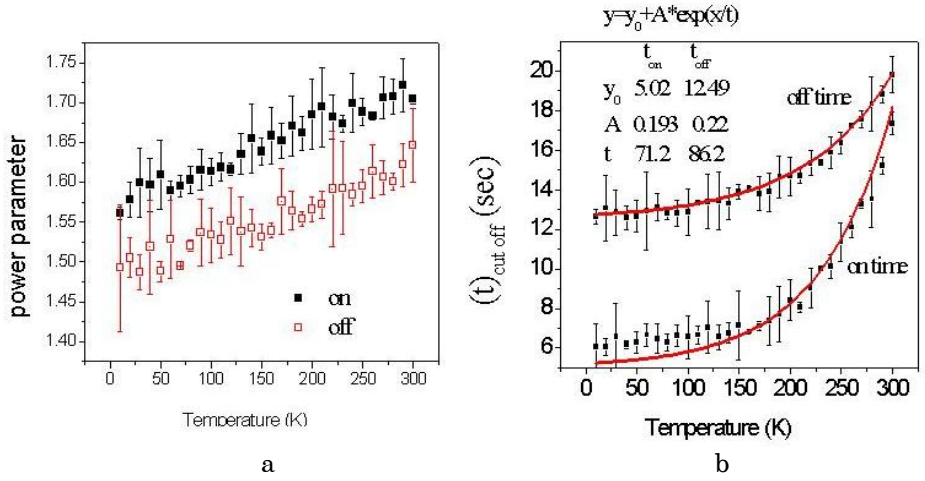

Fig. 4 – m_{on} *and* m_{off} *as a function of temperature* (*a*). m_{on} *was observed to be consistently greater than* m_{off} *and Cut off time for both on and off time distributions as a function of temperature* (*b*). *Both the curves were fitted with exponential curve and the parameters are mentioned*

We have quantified this by noting the time at which the curves (Figure 3) starts to deviate from the straight line. We call this time as the cutoff time. Smaller the cutoff time, larger is the deviation. Figure 4b is the plot of cutoff times corresponding to on and off time distribution as a function of temperature. Both of them could be fitted with an exponential curve, whose parameters are mentioned in the plot. This clearly indicates that the influence of built in electric field due to charge separation on the random walk is to make the on and off time statistics Gaussian like. This can be due to the fact that under an electric field the long steps, which are the signature of Levy random walk, away from the QD reduce, thus the statistics approaches Gaussian. There is some evidence of this behavior in

literature. Shimizu et al. [9] demonstrated experimentally and Sharma et al. [13] showed numerically that the cutoff time of the "on" time distribution becomes smaller when the excitation intensity increases. It should be kept in mind that increasing light intensity increases the built in electrostatic field between electrons and ionized QD, thus induces the same effect as the decreasing temperature, so far as bias in the random walk is considered. Furthermore, it could be seen that cut off time for "off" states are relatively insensitive to the temperature, though a clear temperature dependent is observable. Further investigation is needed to explain this asymmetry in cut off times for "on" and "off" states.

5. CONCLUSION

In summary, we have simulated the temperature dependence of blinking statistics of QD under continuous excitations. With increase in temperature, both m_{on} and m_{off} are found to increase, though m_{on} was consistently larger than m_{off}. At higher time scale, deviation from linearity was observed, which was attributed to the electric field induced suppression of Levy type random walk and is more effective at lower temperature. Asymmetry in temperature dependence of cut off times for "on" and "off" states was observed, which could not be explained at present.

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