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Statistical properties of single molecule under stochastic gating

Bai-Ping Han[a], Li-Shan Gu[a,b], Zi-Wu Ji[a] and Yu-Jun Zheng[a,*]

[a] *School of Physics, Shandong University, Jinan 250100, China*
[b] *Department of Physics, Changji University, Xinjiang 831100, China*

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Abstract. We discuss the blinking statistical behaviors of dynamics of single molecule system, using the recently developed generating function method. We make a thorough study for the fluorescence blinking behaviors and get the statistical properties of the jumping events respectively onto ON state or OFF state, including the waiting time and waiting time distribution for every directional jumping event, the cumulants of jumping events, the cross correlation, the joint probabilities between two directional jumping events and the probabilities.

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Key words: single molecule, stochastic gating, statistical properties

1 Introduction

The observation of blinking phenomenon is ubiquitous for all single molecule studies, such as various quantum dots [1–3], single polymer segment [4],fluorescent proteins [5–8], single dye molecules [9] and so on. About the distribution of sojourn time on ON state (fluorescence) or OFF state (nonfluorescence), there are some descriptions, such as the single exponential distribution for the three-level system, the nonexponential distribution for the condensed phase system with continuum manifold states and power law distribution for the single semiconductor quantum dots. The studies of physical mechanism behind the fluorescence intermittency in quantum dots have been proposed, including different pictures [10].

Due to the effect of conformational (environment) fluctuation, the conversion rate between the ON state and OFF state can be considered as the stochastic variable. In a widely discussed example, i.e., the reaction of ligand binding to the protein, the rate of

*Corresponding author. *Email address:* yzheng@sdu.edu.cn (Y. J. Zheng)

ligand binding to proteins is more slower than that while reaction were completely diffusion controlled. The entrance of ligand to the protein is blocked by a number of side chains and thus ligand could not bind if these side chains were fixed at their equilibrium positions [8]. The blocking behaviors of the side chains act as a gate and the gate would fluctuate between open and closed positions. The opening and closing of the gate is a stochastic process. Further, Zwanzig [7] considered that the conversion process is assumed to be passage through a bottleneck, which is geometrical. The decay rate of passage through the bottleneck is proportional to the area of the bottleneck and the external fluctuations influence the cross-sectional area.

In this study, we assume that the reactivity fluctuates stochastically and the ligand has a finite size. When the radius of bottleneck is smaller than the size of ligand, the decay rate of single molecule is zero, while lager than the size of ligand, the rate of passage through the bottleneck is proportional to the cross-sectional area of this bottleneck. The influence of environment changes the cross-sectional area of bottleneck.

Similarity, the blinking behaviors of an enzyme single molecule can be considered using this stochastic gating model. The reductive and oxidative reactions are respectively corresponding to the activity (ON state) and nonactivity (OFF state) of fluorescence [9].

We use the generating function method to consider the statistics of jumping events, the different directions jumping statistics can be got respectively. The paper is organized as follows. In Section 2 of this paper, we present the theoretical derivation of the generating function for single molecule fluorescence blinking and the statistical quantities, that can be extracted. In Section 3, we give the numerical results of statistical properties for single system fluorescence blinking. The conclusions are given in Section 4.

2 Theoretical framework

The dynamics of fluorescence intermittency is corresponding to the transformation between the ON state and the OFF state in blinking statistics. Due to the influences of environment fluctuations, the transformation rate can be considered as the stochastic variable. The time-dependent dynamics equation about the single biological system can be shown

$$ON \underset{k_{ON}(t)}{\overset{k_{OFF}(t)}{\rightleftarrows}} OFF\,. \tag{1}$$

The generating function approach has been formed to calculate SMS fluorescence blinking statistics behaviors. We have used the generating function to study the statistical properties of single molecule system [11–17]. This approach is amenable to both analytical and numerical calculations for many of statistical properties inherent to SMS measurement. In comparison with previous works [18], we introduce two "auxiliary" variables s_1 and s_2, respectively accounting the OFF state jumping times and ON state jumping times within a time interval. The generating function form about Eq. (1) was