J. At. Mol. Sci.
doi: 10.4208/jams.052516.081516a

Vol. 7, No. 4, pp. 191-199
November 2016

Photon statistical properties emitted from single molecule derived from generating function method

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Received 25 May 2016; Accepted (in revised version) 15 August 2016
Published Online 10 November 2016

Abstract. In this paper, we summarize the photon emission statistical properties of single molecule system, using the recently developed generating function method. Through the introduce of one or double "auxiliary" variables, we can investigate the first moment and second moment of photon emission statistics driven by different external fields, such as the line shapes and Mandel's Q parameters, the photon emission probabilities, the probability distribution of time between successive emission, the waiting time and the waiting time distribution, the cross correlation, the joint probabilities, etc. Among,the first moment of photon statistics and the experimental results of optical amplification are in good agreement.

PACS: 33.50.Dq, 42.50.Ar

Key words: Two-Level System, Generating Function, Photon Counting Statistics.

1 Introduction

Single molecule technology has been used for the research of condensed phase systems in multiple areas [1–5] and has developed rapidly for the several decades. There are a large number of experimental studies [6–10]. Correspondingly, there are several developed theoretical methods [11–15] about single molecule fluorescence photon, including Master equation method, Wiener-Khintchine method, quantum jumping method, generating function and Levy-walk method and so on. Among, the generating function method has studied many single molecule systems and gives some photon emission statistical properties [16–26]. Also, this method can get the high-order moments, the bunching and anti-bunching effect of single molecule emission photon distribution.

We study the photon emission of this dissipative two-level system under different external fields and give the photon emission statistical properties of single molecules

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system, using the recently developed generating function method. First of all, the time-dependent Hamiltonian of single molecule under pump-probe field can be written as:

$$\mathcal{H}(t)=\hbar\omega_g|g\rangle\langle g|+\hbar\omega_e|e\rangle\langle e|-\mu_{ge}\cdot E(t)(|g\rangle\langle e|+|e\rangle\langle g|). \tag{1}$$

The external field can be expressed as [27], and has a variety of forms. The different forms will be displayed in the flowing, corresponding to the related figures. The dynamical evolution of density element satisfies the quantum Liouville-Von Neumann equation $\partial\rho/\partial t=-i[H,\rho]/\hbar$ [1,28]. The density matrix elements have four parts $\rho_{ee}(t)$,$\rho_{gg}(t)$,$\rho_{ge}(t)$ and $\rho_{eg}(t)$. The above equations can be rewritten as $\dot{\rho(t)}=L_0(t)\rho(t)+L_1(t)\rho(t)$. Within the optical Bloch frame, we can resolve the density operator into the manner $\rho(t)=\sigma^{(0)}(t)+\sigma^{(1)}(t)+\sigma^{(2)}(t)+\cdots$. Here $\sigma^{(n)}(t)=(\sigma^{(n)}_{ee}(t),\sigma^{(n)}_{gg}(t),\sigma^{(n)}_{ge}(t),\sigma^{(n)}_{eg}(t))^\dagger$. The $\sigma^{(n)}(t)$ is the part of the density matrix and is corresponding to n photons have been emitted. So, we can defined the generating functions as

$$\begin{aligned}G_{ee}(s,t)&\equiv\sum_{n=0}^{\infty}\sigma^{(n)}_{ee}(t)s^n;\\G_{gg}(s,t)&\equiv\sum_{n=0}^{\infty}\sigma^{(n)}_{gg}(t)s^n;\\G_{ge}(s,t)&\equiv\sum_{n=0}^{\infty}\sigma^{(n)}_{ge}(t)s^n;\\G_{eg}(s,t)&\equiv\sum_{n=0}^{\infty}\sigma^{(n)}_{eg}(t)s^n.\end{aligned} \tag{2}$$

In the rotating wave approximation (RWA), we derive the evolution differential equations of the generalized Bloch vectors $\mathcal{U}(s,t)$, $\mathcal{V}(s,t)$, $\mathcal{W}(s,t)$, and $\mathcal{Y}(s,t)$ as

$$\begin{aligned}\mathcal{U}(s,t)&\equiv\frac{1}{2}(\mathcal{G}_{ge}e^{-i\omega_L t}+\mathcal{G}_{eg}e^{i\omega_L t}),\\\mathcal{V}(s,t)&\equiv\frac{1}{2i}(\mathcal{G}_{ge}e^{-i\omega_L t}-\mathcal{G}_{eg}e^{i\omega_L t}),\\\mathcal{W}(s,t)&\equiv\frac{1}{2}(\mathcal{G}_{ee}-\mathcal{G}_{gg}),\\\mathcal{Y}(s,t)&\equiv\frac{1}{2}(\mathcal{G}_{ee}+\mathcal{G}_{gg}).\end{aligned} \tag{3}$$

Following, we enumerate the photon emission statistical properties of single molecules system, Such as the line shapes and Mandel's Q parameters, the photon emission probabilities, the probability distribution of time between successive emission, the waiting time and the waiting time distribution, the cross correlation, the joint probabilities.