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Emission spectrum of two-level single molecule *via* photon counting statistics

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Abstract. In this paper, we introduce a new way to obtain the emission spectrum of single molecule driven by external field. A virtual probe field is introduced to simulate the vacuum field, which causes the spontaneous emission phenomena of the excited single molecule. The statistical properties of the emission photons caused by the virtual probe field could be used to obtain the emission spectrum of the single molecule system. The results demonstrate the well-known Mollow triplet splitting phenomenon as the single molecule is driven by strong external field.The abstract should provide a brief summary of the main findings of the paper.

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Key words: Two-Level System, Emission Spectrum, Generating Function.

1 Introduction

The spectrum of the single molecule could be used to directly observe the change of the state of the system. The well-known one is the Autler-Townes splitting [1], which was first observed by Autler and Townes. Generally, a single quantum system (including single atoms, single molecules, and single quantum dots, *etc.*) driven by strong external field, the coupling states to the external field of the single quantum system could be splitting into two sub-states, this is the well known Autler-Townes splitting phenomenon. It has been studied by several groups [2–5]. This splitting reflects in the emission spectrum is also a well known phenomenon — Mollow triplet splitting [6], which was predicted theoretically by Mollow, then several groups have observed the Mollow triplet splitting phenomenon in experiment [7, 8].

Generally, the emission spectrum is considered as a Fourier transformation of the time dependent dipole-dipole correlation [6, 9], which reflect the emission intensity distribution of the emission photon frequency. On the other way, one could directly obtain

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the emission intensity distribution of the emission photon frequency *via* counting the emission photons at frequency ω [10]. As we all know, the spontaneous emission of an excited quantum system is caused by the vacuum field [11,12], and the vacuum field include all frequency modes [12]. That means we can not get a the frequency distribution of the emission photons directly from the vacuum field. If we introduce a virtual, changeable frequency probe field to simulate a frequency mode of the vacuum field, then one can get the emission photon number at a fixed frequency. By scanning all the frequency modes, one could obtain the emission intensity distribution versus the emission photon frequency ω, and directly obtain the emission spectrum.

In this paper, we demonstrate how to obtain the emission spectrum of a two-level single molecule driven by external field, the generating function approach is employed to obtain the emission intensity (or average emission photons, which is proportional to the emission intensity at fix time t) at frequency ω. The generating function approach is well used to discuss the emission photon counting statistical properties [13–22]. It is also used to discuss the emission spectrum of driven single molecule [10]. In Ref. [10] they directly analyze the emission photons, have gotten the emission spectrum and the corresponding Mandel's Q parameter.In this paper, we introduce a new way to obtain the emission spectrum *via* introducing a virtual probe field. The Mandel's Q parameter in this paper is corresponding to the emission photon caused by the probe field, it is different compared with that in the Ref. [10].

This paper organized as follows: In Sec. 2 we briefly review the generating function approach of single molecule for photon counting statistics, and apply the generating function approach to obtain the emission photon number $\langle N\rangle$ and the corresponding Mandel's Q parameter. The numerical results and discussions are demonstrated in Sec. 3. Our conclusions will be presented in Sec. 4.

2 Theory

The Hamiltonian of a two-level single molecule system driven by external field (including a pump field and a virtual probe field), can be written as

$$\mathcal{H}=\mathcal{H}_s+\mathcal{V}_{se}+\mathcal{V}_{sp}, \tag{1}$$

where $\mathcal{H}_s$ is the Hamiltonian of the "bare" two-level system, and the $\mathcal{V}_{se}$ is the interaction between the two level system and the pump field, $\mathcal{V}_{sp}$ is the interaction between the two level system and the probe field. The Hamiltonians can be expressed as

$$\begin{aligned}
&\mathcal{H}_s=\hbar\omega_a|a\rangle\langle a|+\hbar\omega_b|b\rangle\langle b|,\\
&\mathcal{V}_{se}=\hbar\Omega\cos(\omega_L t)(|a\rangle\langle b|+|b\rangle\langle a|),\\
&\mathcal{V}_{sp}=\hbar\Omega_p\cos(\omega_p t)(|a\rangle\langle b|+|b\rangle\langle a|),
\end{aligned} \tag{2}$$

where $\Omega\equiv-\boldsymbol{\mu}_{ab}\cdot\mathcal{E}_0/\hbar$, and $\Omega_p\equiv-\boldsymbol{\mu}_{ab}\cdot\mathcal{E}_p/\hbar$ are the Rabi frequencies of the pump field and probe field, respectively, the $\boldsymbol{\mu}_{ab}$ is the transition dipole moment of the ground state