Effect of Background Noise on the Photon Statistics of Triggered Single Molecules *

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We theoretically derive exact expressions for Mandel's Q parameter of the triggered single molecular source, which is inferred from the probabilities $P_{RS}(n)$ using the recorded of each photon detection event based on Hanbury Brown and Twiss detection. The real triggered source is recognized as an ideal single photon source with a Poissonian statistics background. How to decease the background and to increase the efficiency are discussed. It is established that the sub-Poissonian statistics formation can be determined by comparing the measured Q_{RS} of the real single triggered molecular with Q_C of the Poissonian source containing the same mean photons. By this method, we also give an efficient way to measure signal-to-background ratios of triggered single photons.

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The development of efficient single photon sources is an important prerequisite for quantum information processing in the forms of quantum computing,[1] quantum cryptography,[2] quantum teleportation,[3,4] and quantum networking.[5] The recent experimental demonstration of some solid-state triggered single-photon devices including molecules[6−8] and semiconductor quantum dots[9] have shown an ability to emit photons one by one. In an ideal single-photon emitter, one can moreover hope to generate a regulated photon stream, containing one and only one photon in a given time interval. Such a source would then be virtually free of intensity fluctuations, therefore corresponding to perfect intensity squeezing.

Photon statistics for single molecules are normally measured using the second-order correlation function $g^{(2)}$ based on the standard Hanbury Brown and Twiss (HBT) detection, as explained in Refs. [10,11], whereas the time intervals of photon events are measured and the number of arriving photons within a certain counting time is measured. Nevertheless, this technique can hardly be used to extract the intensity fluctuations on time scale longer than a single pulse duration. It is therefore chosen to record each photon-detection event.[8] As presented in Ref. [8], the set of time tagged photoncounts allows one to make a direct time domain fluctuation analysis. From this, one can deduce all moments of the photon statistics in order to measure Mandel's Q parameter of a triggered molecule source. In addition, Q parameters for weak coherence pulse photon statistics were measured with the excess noise of the laser diode in the range of 0–15 dB.[12] It appeared in the work that the excess noise dependence arises as a main factor in Q parameter measurement.

Due to remnant Raman scattering, fluorescence of the substrate or of the detection optics, a photon from the background which triggers a coincidence with a photon from the molecule (or from the background) should seriously limit the practical use of such a photon source.[13] Another kind of background noise is the dark counting coming from single photon detectors. Nevertheless, all of the background can be regarded as the following Poissonian statistics.[10] In this Letter, we derive exact expressions for the Mandel parameter of the triggered single molecular source, which is inferred from the probabilities $P_S(n)$ for the source, of measuring $n = 0$, 1, 2 photoncounts per excitation pulse, using the recorded of each photon detection event. To our knowledge, the effects of background noise on the photon statistics of triggered single molecules have not being discussed in this way.

We first divide the finite time interval from t to $t + T$ into a large number of differential intervals of width delta t. The random variable W occurring within the finite time interval t to $t + T$ is then obtained by adding together the averages for all the differential intervals, and this clearly yields

$$W \equiv \varepsilon \int_t^{t+T} I(r, t')dt'. \qquad (1)$$

Here $I(r, t')$ is the instantaneous light intensity, r is

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the position of the detector; and ε is a constant characterizing the detector efficiency for a particular frequency and a particular polarization.

The photon number distribution can be expressed as

$$P(n,t,T) = \left\langle \frac{1}{n} W^n e^{-W} \right\rangle. \tag{2}$$

In order to quantify the fluctuations of the number of photons n detected per pulse, an important figure of merit is the Mandel parameter:[13]

$$Q \equiv \frac{\langle (\Delta n)^2 \rangle}{\langle n \rangle} - 1. \tag{3}$$

Here $\langle n \rangle$ is the average number of detections occurring within the finite time interval t to $t+T$.

Poissonian photoncounts statistics yields $Q = 0$ whereas the sub-Poissonian and the super-Poissonian statistics correspond respectively to the negative and positive Q.

In order to identify rapidly triggered single molecule emission, a standard HBT setup is usually used to split the beam and detect single photons on two identical single-photon-detectors, such as avalanche photon diode (APD), PMT or CCD. However, since each detection is affected by the dead time[14] (about *ns* level) of the detectors, if the fluorescence lifetime and the pump laser pulse duration are much less than the dead time, each detector cannot detect more than one photon per pulse.

For a coherent state of light containing α photon per excitation pulse, the photon number probability is $P(n) = a^n \times e^{-\alpha}/n!$,[15] one can readily calculate the counting probability distribution:

$$P_C(0) = e^{-\alpha}, \tag{4}$$

$$P_C(1) = 2e^{-\alpha/2}(1 - e^{-\alpha/2}), \tag{5}$$

$$P_C(2) = (1 - e^{-\alpha/2})^2, \tag{6}$$

and $\bar{n} = P_C(1) + 2P_C(2) = 2(1 - e^{-\alpha/2})$, where $\bar{n}$ is the mean number of detected photons per pulse.

One predicts a value

$$\begin{aligned} Q_C =& \frac{1}{\bar{n}}[P_C(0) \times \bar{n}^2 \\ &+ P_C(1) \times (\bar{n}-1)^2 + P_C(2) \times (\bar{n}-2)^2] - 1 \\ =& e^{-\alpha/2} - 1 = -\bar{n}/2. \end{aligned} \tag{7}$$

We find that, due to the photondetection dead time, the triggered reference source also yields a sub-Poissonian counting statistic.

In addition, in the case of an ideal single photon source (SPS), we assume that η is the quantum efficiency of the detectors,

$$P_S(0) = 1 - \eta, \tag{8}$$

$$P_S(1) = \eta, \tag{9}$$

$$P_S(n, n > 1) = 0, \tag{10}$$

and therefore

$$Q_s = -\eta. \tag{11}$$

A real triggered single molecule (RTSM) is recognized as an ideal SPS with a Poissonian statistics background noise. We assume the background noise with the mean number γ of photons per pulse. One can readily calculate the counting probability distribution and show that

$$P_{RS}(0) = (1-\eta)e^{-\gamma}, \tag{12}$$

$$P_{RS}(1) = \eta e^{-\gamma/2} + (1-\eta) \times 2e^{-\gamma/2}(1 - e^{-\gamma/2}), \tag{13}$$

$$P_{RS}(2) = \eta(1 - e^{-\gamma/2}) + (1-\eta)(1 - e^{-\gamma/2})^2. \tag{14}$$

The mean number of detected photons per pulse

$$\begin{aligned} \bar{n}_{RS} &= P_{RS}(1) + 2P_{RS}(2) \\ &= 2(1-\eta)(1 - e^{-\gamma/2}) + 2\eta - \eta e^{-\gamma/2}. \end{aligned} \tag{15}$$

One predicts RSTM's Mandel parameter

$$\begin{aligned} Q_{RS} =& -[2\eta e^{-\gamma/2} - 4e^{-\gamma/2} + 2e^{-\gamma} - 2\eta e^{-\gamma} \\ &+ \eta^2 e^{-\gamma} + 2][2 - 2e^{-\gamma/2} + \eta e^{-\gamma/2}]^{-1}. \end{aligned} \tag{16}$$

Moreover, to the Poissonian coherent source (C) containing the same mean number $\bar{n}_{RS}$ of photons per pulse, the Mandel parameter

$$Q_c = -\frac{\bar{n}_{RS}}{2} = \frac{\eta e^{-\gamma/2}}{2} - \eta - (1-\eta)(1 - e^{-\gamma/2}). \tag{17}$$

A number of features of the simple solution are worth pointing out:

(a) Unlike the fundamental Mandal parameter $Q = 0$, here we present the effective Mandel parameter $Q_e = -\bar{n}/2$ of Poissonian photoncounts statistics, with $\bar{n}$ being the mean photon numbers based on the HBT detection. Sub-Poissonian and super-Poissonian statistics should correspond respectively to the values smaller and larger than Q_e.

(b) From Eqs. (13) and (14), one can infer η and γ using the measured $P_{RS}(1)$ and $P_{RS}(2)$. The signal-to-background ratios (SBR) η/γ could be calculated easily. In this way we give an efficient method to measure SBR of triggered single photons. Accordingly, SBRs are normally measured by using the ratio of the area of the central peak (proportional to $(B^2 + 2SB)$) to the lateral peak (proportional to $(B+S)^2$) as explained by Lounis and Moerner[11] with the normally start-stop technology.

(c) In the case of an ideal SPS, we have $Q_s = -\eta$, with the same value predicted in Ref. [16]. Unlike the Poissonian statistics, there is not $P(n, n > 1)$ counting probability distribution for the ideal SPS, and the dead time does not affect the detection result.

From Eqs. (16) and (17), we introduce a normalized relative contrast R defined, on the sample, by

$$R = (Q_{RS} - Q_C)/Q_{RS}. \tag{18}$$

In the experiment, it is however difficult to compare Q_{RS} with Q_C if the relative contrast R is less than 0.1 considering the measurement error.[8,12]

Figure 1 shows the Mandel parameter Q_{RS} for the RTSM, Q_C for the Poissonian source and relative contrast R as a function of the background noise with mean photons number γ and the quantum efficiency $\eta = 0.0445$, which is the typical parameter from Ref. [8]. We find that the relative contrast R becomes approximately zero with the increasing background noise.

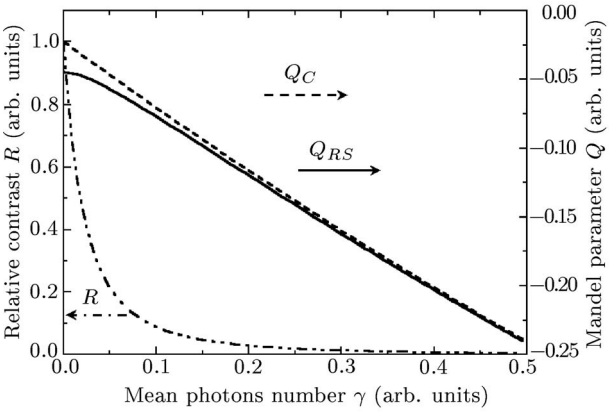

Fig. 1. Mandel parameter Q_{RS} for an RTSM [solid line, Eq. (16)], Q_C for the Poissonian source [dashed line, Eq. (17)] and relative contrast R [dot-dot-dashed line, Eq. (18)] as a function of mean photons number γ, with quantum efficiency $\eta = 0.045$.

As higher background may induce inefficient measurement, it is necessary to reduce the background noise. In practice, selecting a special polymer for the single molecules may reduce the background noise. For example, Fleury *et al.*[10] and Lounis and Moerner[11] recently observed antibunching in the fluorescence of a remarkably photon-stable system made from terrylene molecules embedded in the crystalline host of para-terphenyl. They obtained SBR nearly to be 5, However, In Roch's experiment,[17] the dye (terrylene) molecules are embedded in a highly transparent polymer film of polymethyl methacrylate (PMMA) and the SBR was increased to 20.

Another efficient way to reduce the background is to select low-dark-counting single photon detectors.

Figure 2 shows the Mandel parameter Q for the RTSM, Q_C for the Poissonian source and relative contrast R as a function of the quantum efficiency η, calculated with $\gamma = 0.0022$, which is from the typical parameter from Ref. [8]. To a particular background noise, the intensity statistics are close to a Poisson law with smaller quantum efficiency η.

In early SPSs operated at cryogenic temperature,[18] the collection efficiency of photons is barely higher than a few 10^{-3} in these experiments. Because of this far small efficiency, the intensity statistics are very close to a Poisson law at the place where the stream of photons is available.

Another route is to realize RTSMs at room temperature.[10,11,17] In this case, higher collection efficiency (around 5%) is achieved. One can still improve the detection efficiency simply by covering the sample with a silver layer mirror[17] or microcavity.[19]

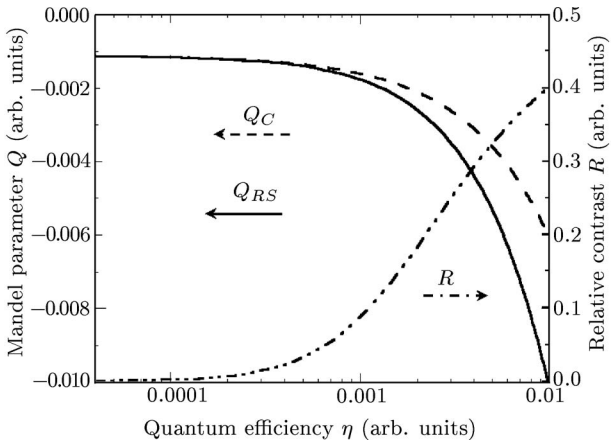

Fig. 2. Mandel parameter Q_{RS} for an RTSM [solid line, Eq. (16)], Q_c for the Poissonian source [dashed line, Eq. (17)] and relative contrast R [dot-dot-dashed line, Eq. (18)] as a function of the quantum efficiency η, calculated with mean photons number $\gamma = 0.0022$.

In summary, as an RTSM being recognized as an ideal SPS with a Poissonian statistics background noise, we have theoretically derived exact expressions of Mandel's Q parameter based on HBT detection. The effective Mandel parameter $Q_e = -\bar{n}/2$ of Poissonian photoncounts statistics is given out. It is presented that the set of time tagged photoncounts allows one to measure Mandel's Q parameter. One can infer η and γ using the measured $P_{RS}(1)$ and $P_{RS}(2)$. In this way we give an efficient method to measure SBR of triggered single photon. In particular, we have discussed the effect of background on Mandel's Q parameter.

This analysis can be straightforwardly applied to characterize SPSs. It is also complementary to fluorescent correlation spectroscopy technique for investigating photochemical properties at the single molecule level as well as of single molecules in condensed phases.[20]

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References

[1] Turchette Q A, Hood C J, Lange W, Mabuchi H and Kimble H J 1995 *Phys. Rev. Lett.* **75** 4710
[2] Beveratos A, Brouri R, Gacoin T, Villing A, Poizat J P and Grangier P 2002 *Phys. Rev. Lett.* **89** 187901
[3] Bennett C H, Brassard G, Crepeau C, Jozsa R, Peres A and Wootters W K 1993 *Phys. Rev. Lett.* **70** 1895
[4] Furusawa A, Sorensen J L, Braunstein S L, Fuchs C A, Kimble H J and Polzik E S 1998 *Science* **282** 706
[5] Kuhn A, Hennrich M and Rempe G 2002 *Phys. Rev. Lett.* **89** 067901
[6] Basche T, Moerner W E, Orrit M and Talon H 1992 *Phys. Rev. Lett.* **69** 1516
[7] Brunel C, Lounis B, amarat P T and Orrit M 1999 *Phys. Rev. Lett.* **83** 2722
[8] Treussart F, Alleaume R, Le Floch V, Xiao L T, Courty J M and Roch J F 2002 *Phys. Rev. Lett.* **89** 093601
[9] Santori C, Pelton M, Solomon G, Dale Y and Yamamoto Y 2001 *Phys. Rev. Lett.* **86** 1502
[10] Fleury L, Segura J M, Zumofen G, Hecht B, and Wild U P 2000 *Phys. Rev. Lett.* **84** 1148
[11] Lounis B and Moerner W E 2000 *Nature* **407** 491
[12] Xiao L T, Alleaume R, Dinh-Xuan Q, Treussart F, Journet B and Roch J F 2003 *SPIE* **4986** 58
[13] Michalet X and Weiss S 2002 *C. R. Physique* **3** 619
[14] Overbeck E, Sinn C, Flammer I and Ricka J 1998 *Rev. Sci. Instrum.* **69** 3515
[15] Brattke S, Varcoe B and Walther H 2001 *Phys. Rev. Lett.* **86** 3534
[16] Abate J, Kimble H and Mandel L 1976 *Phys. Rev.* A **14** 788
[17] Treussart F, Clouqueur A, Grossman C and Roch J F 2001 *Opt. Lett.* **26** 1504
[18] Kim J, Benson O, Kan H and Yamamoto Y 1999 *Nature* **397** 500
Brunel C, Lounis B, Tamarat P and Orrit M 1999 *Phys. Rev. Lett.* **83** 2722
[19] Moreau E, Robert I, Gerard J M, Abram I, Manin L and Thierry-Mieg V 2001 *Appl. Phys. Lett.* **79** 2865
[20] Pan X Y, Jiang H B, Liu C L, Gong Q H, Zhang X Y, Zhang Q F, Xu B X and Wu J L 2003 *Chin. Phys. Lett.* **20** 133