Probing photon energy statistics of an emitter using photon correlations

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We investigate the possibility of measuring the homogeneous and inhomogeneous contribution to the linewidth of a spectrally diffusing single photon emitter using a simple photon correlation spectroscopy method (PCS). The photon energy statistics of the homogeneous line (poissonian) and of the spectral diffusion (first order markovian) being of different natures, they act differently on the half-line autocorrelation function (HLAF). We model here their effects and show it is possible to extricate them, opening the opportunity to determine separately the homogeneous linewidth, and the spectral diffusion amplitude.

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I. INTRODUCTION

It is known since the pioneer works of Handbury-Brown and Twiss on the correlation of an optical field \cite{1}, and of Kimble, Dangennais and Mandel on the correlation of a quantum state of the light \cite{2}, that it is possible to probe the emission statistics of an emitter by photon correlations. In particular Kimble et. al showed that for a two levels system, the autocorrelation function of the emitted optical field increases with the delay $\tau$ \cite{3}. This is a characteristic of a single photon emission. In condensed matter systems, the energies of the emitted single photons can fluctuate. The interaction of the emitter with the numerous degrees of freedom of the surrounding environment leads to random fluctuations of the emission energy. Since quantum computation requires indistinguishable photons \cite{1} in order to proceed to efficient two-photon interferences \cite{5}, this is a severe limitation in the use of solid state systems for such operations. This dephasing phenomenon is the result of several different processes. From the energy statistics point of view, one can divide them in two categories: Poissonian processes, such as exciton-phonon coupling, randomly affect the energy of the emitted photons. They are "memoryless" processes as the energy of the emitted photon at time $t$ does not depend on the energy of the emitted photon at time $t-\tau$ whatever the value of $\tau$. We define the energy distribution of these uncorrelated random processes as the homogeneous linewidth of the emitter. On the other hand, a correlated process can shift the emitter energy. Fluctuations of the electronic environment are also a source of dephasing. This random Stark-shift of the emission energy is called spectral diffusion \cite{6}. In opposition with phonon coupling, spectral diffusion is time-correlated and it is a reasonable approximation to consider it as a first order Markovian process with a correlation time $\tau_c$ \cite{7,8}. Unlike the poissonian process, which is independent of its history, the first order markovian process is influenced by its immediate or most recent past. Thus, the energy of the emitted photon shifted by the spectral diffusion depends on the energy of the previously emitted photon. In a previous publication \cite{9}, it was shown that, by taking the Handbury-Brown and Twiss setup and by adding detection energy conditions, such as selecting only one half of the emission line, one has access to the subnanosecond correlation time of the spectral diffusion $\tau_c$ \cite{9}. In this letter, we show that this measurement technique is even more powerful since the resulting half line autocorrelation function (HLAF) brings informations on the relative contribution of the correlated and uncorrelated processes. Thus, such a slight modification of the original HBT setup, which was providing emission statistics of the emitter, gives as well access to the energy statistics of the emitter. In a first part, we describe the main principles of the experiment. Then, after showing that it is always possible to separate the emission statistics and the energy statistics contributions on the HLAF, we derive analytically the HLAF of a spectrally diffusing single photon emitter with a finite homogeneous linewidth. After discussing its properties, we confirm the validity of our model with a Monte Carlo simulation, building numerically the HLAF of a spectrally diffusing poissonian emitter and simulating the influence of a finite homogeneous linewidth. To finish, we highlight the possible direct application of this theoretical result: the separate determinations of the homogeneous linewidth and the spectral diffusion amplitude.

II. PRINCIPLE OF THE MEASUREMENT

The principle of the PCS experiment is to convert fluctuations of the emitter energy in intensity fluctuations. In fig. 1 a) we consider the situation where all the photons are collected and sent to the HBT setup whatever their energies (photons coming from the right side of the line are coloured in red, from the left side in blue). The scheme in fig. 1 b) represents the single photon emission with photons arriving one by one with a characteristic time defined by the emitter radiative lifetime. The HBT setup allows to measure the intensity first order correlation function \cite{10}:
\[ g^2(\tau) = \frac{\langle (I(0)I^*(\tau)) \rangle}{\langle I(\tau) \rangle^2} \]  

(1)

This function is plotted in fig. 1 c). The dip at zero delay is the signature of single photon emission. It is non-null at zero delay only because of the convolution with time resolution of the photodetectors.

We introduce now a detection energy condition such as we only detect photons emitted from the high energy side of the spectrum called right side (R side) in fig. 2. Therefore, we can discriminate the photons emitted from the high energy side of the spectrum (ie. R side, red spikes in fig. 2 b) ) from the photons emitted in the low energy side (L side, blue spikes in fig. 2 b) ). In the fig. 2 b), bunches of single photons (red spikes) emitted from the R side are observed because the spectral diffusion is a time-correlated process (ie. with memory). On the HLAF (fig. 2 c)), it results in a bunching combined with a zero delay dip due to the subpoissonian nature of the emission. If we consider that the emitter homogeneous linewidth is infinitely small, the HLAF can be easily calculated with a rate equation model by considering a split two-levels system with a probability for the emitter energy to switch from one side to the other one.

\[ \text{HLAF}(\tau) = g^2(R, R, \tau) = [1+exp(-\gamma_c \tau)][1-exp(-(r+\gamma)\tau)] \]  

with \( \gamma_c = \frac{1}{\tau_c} \), the correlation rate, \( r \) the pump rate and \( \gamma \) the spontaneous emission rate.

It is the product of two expressions. Since \([1 - exp(-(r + \gamma)\tau)]\) describes the single photon behavior of the emitter, \([1 + exp(-\gamma_c \tau)]\) describes the spectral diffusion phenomenon and its influence on the ability of the emitter to send photons in the right spectral window. The spectral diffusion term is equal to 2 for the null delay.

We now consider a finite homogeneous linewidth (fig. 3) . The mean value of the homogeneous lineshape transits from one side of the spectral window to the other with the time constant \( \tau_c \). However, even if it is centered in the left side, a photon can be emitted in the right side and vice versa. The consequences on the photons statistics are represented in fig. 3 b). To the bunching statistics due to the time correlated spectral diffusion process it has to be added an uncorrelated random distribution of the photons on both spectral windows, degrading the photons "bunching". The consequence on the HLAF is a diminution of the bunching contrast, as the energies of photons are less time correlated (fig. 3 c)). The importance of this contrast is linked to the relative importance of the homogeneous linewidth compared to the spectral diffusion amplitude. To understand this dependence, we calculate in the following the HLAF corresponding to a spectrally diffusing single photon emitter with a finite homogeneous linewidth.

### III. ENERGY AND EMISSION SEPARATION

The aim of this section is the obtention of a preliminary result, which will simplify the calculations leading to the derivation of the HLAF for a non-negligible homogeneous linewidth.

We propose to show that the HLAF is the product of the spectral diffusion part (carrying information on the correlation of the emitter energy) and the single photon part (carrying information on the correlation of the emission). This result only holds when emission and energy position of the line are independent. The emitter is seen as a two-level system (TLS) of fluctuating energy.

Let us define the following ensembles:
two independent events, so:

\[ \rho(\gamma_t^\tau \cap \gamma_t^0) = \rho(\gamma_t^\tau)\rho(\gamma_t^0) \] (3)

We go back to the expression of autocorrelation function in term of probabilities:

\[ g^2(\tau) = \frac{\rho(\gamma^\tau \mid \gamma_0^0)}{\rho(\gamma^0)} \] (4)

To detect a photon at time \( t \), one needs to have a photon emitted by the TLS at time \( t \) and to have the TLS energy in the right spectral window at time \( t \):

\[ \gamma_t^\tau = \gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma 0} \] (5)

Therefore,

\[ \rho(\gamma^\tau \mid \gamma_0^0) = \rho(\gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma 0} \mid \gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma 0}) \] (6)

Using \[ (5) \] in \[ (6) \], we have:

\[ \rho(\gamma(\tau) \mid \gamma(0)) = \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0} \cap \gamma_{\tau}^{\gamma 0} \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0} \cap \gamma_{\tau}^{\gamma 0}) \]
\[ = \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0})\rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0}) \] (7)

We can now find the general expression, separating the TLS emission and energy correlation functions in the HLAF:

\[ g^2(\tau) = \frac{\rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0})\rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0})}{\rho(\gamma_{\tau}^{\gamma \tau})\rho(\gamma_{\tau}^{\gamma 0})} = g_{\tau}^{\gamma \tau}(\tau)g_{\tau}^{\gamma 0}(\tau) \] (8)

This result will make our future calculations of HLAF easier, since the emission part \( g_{\tau}^{\gamma \tau}(\tau) \) will always be the same and we will only have to focus on the derivation of \( g_{\tau}^{\gamma 0}(\tau) \), the second order correlation function of the TLS energy position.

IV. HALF-LINE AUTOCORRELATION FUNCTION

Since the mean TLS energy fluctuates from the right side to the other with a correlated statistics, but can also be randomly, and with no time-correlation, distributed in energy along the lorentzian profile imposed by the homogeneous linewidth, The total energy \( E^t \) of the TLS at time \( t \) is the sum of two random variables:

\[ E^t = \mu^t + \epsilon^t \] (9)

where \( \mu^t \) is the energy position of the center of the homogeneous lineshape at time \( t \). It is distributed along the gaussian distribution of the fluctuations \[ [12][13] \]. This random variable is time correlated. It means that the value of the random variable at time \( t + \tau \) is influenced by its value at time \( t \) and:

\[ \langle \mu^t \mu^{t+\tau} \rangle \neq \langle \mu^t \rangle \langle \mu^{t+\tau} \rangle \] (10)

\( \epsilon^t \) is the energy shift due to the homogeneous linewidth at time \( t \). We assume it has a lorentzian distribution centered on \( \mu^t \). This variable is described by a poissonian process and is not time-correlated.

\[ \langle \epsilon^t \epsilon^{t+\tau} \rangle = \langle \epsilon^t \rangle \langle \epsilon^{t+\tau} \rangle = \langle \epsilon^0 \rangle \langle \epsilon^\tau \rangle = \langle \epsilon^\tau \rangle^2 \] (11)

The spectral window of detection is defined by the energy interval \( I_n \). In this letter, we consider, \( I_n = [0, +\infty[ \), i.e. corresponding to the R side.

The energy of the TLS is in the right spectral window at time \( t \) when \( E^t \epsilon I_n \), so when:

\[ \langle \epsilon^t + \mu^t \rangle \epsilon I_n \] (12)

Therefore, we have:

\[ \gamma_{\tau}^{\gamma \tau} \equiv \{ (\epsilon^t + \mu^t) \epsilon I_n \} \]
\[ \gamma_{\gamma 0}^{\gamma 0} \equiv \{ (\epsilon^0 + \mu^0) \epsilon I_n \} \]

We also define the following ensembles:

- \( \gamma_{\tau}^{\gamma \tau} \): ensemble of \( \mu \) such as \( \mu^t \epsilon I_n \).

Figure 3. a) spectrum of a spectrally diffusing emitter with a finite linewidth: only the right spectral window is selected for detection. b) photons statistics of the emitter in the PCS experiment: only red photons are detected c) HLAF in the case of a non-negligible homogeneous linewidth

- \( \gamma_{\tau}^{\gamma \tau} \): ensemble of events such as a photon is detected at time \( t \).
- \( \gamma_{\tau}^{\gamma \tau} \): ensemble of events such as a photon is emitted by the TLS.
- \( \gamma_{\tau}^{\gamma \tau} \): ensemble of events such as the TLS energy is in the right spectral window.

The emission of a photon by the two level system (TLS) and its energy situated in the right spectral window are two independent events, so:

\[ \rho(\gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma \tau}) = \rho(\gamma_{\tau}^{\gamma \tau})\rho(\gamma_{\tau}^{\gamma \tau}) \] (3)

We go back to the expression of autocorrelation function in term of probabilities:

\[ g^2(\tau) = \frac{\rho(\gamma^\tau \mid \gamma_0^0)}{\rho(\gamma^0)} \] (4)

To detect a photon at time \( t \), one needs to have a photon emitted by the TLS at time \( t \) and to have the TLS energy in the right spectral window at time \( t \):

\[ \gamma_\tau^\tau = \gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma \tau} \] (5)

Therefore,

\[ \rho(\gamma^\tau \mid \gamma_0^0) = \rho(\gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma \tau} \cap \gamma_{\tau}^{\gamma \tau}) \] (6)

Using \[ (5) \] in \[ (6) \], we have:

\[ \rho(\gamma(\tau) \mid \gamma(0)) = \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0} \cap \gamma_{\tau}^{\gamma 0} \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0} \cap \gamma_{\tau}^{\gamma 0}) \]
\[ = \rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0})\rho(\gamma_{\tau}^{\gamma \tau} \mid \gamma_{\tau}^{\gamma 0}) \] (7)
$-\mu_{\text{out}}^0$: ensemble of $\mu$ such as $\mu_t^i \notin I_n$

We found out in the previous subsection that we can always write down the autocorrelation function as:

$$g^2(\tau) = g_{\text{emp}}^2(\tau)$$

with $g_{\text{emp}}^2(\tau) = \frac{\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0)}{\rho(\gamma_{\text{emp}}^0, \tau)}$.

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho((\{\varepsilon^T + \mu^T\}e(I_n)) | \gamma_{\text{emp}}^0)$$

(13)

We use here the law of total probability which asserts that for an ensemble $A$ and its partition $\{A_i\}$ such as $A = \sum_i A_i$,

$$P(B) = \sum_i P(B | A_i)P(A_i)$$

So, eq. (13) becomes:

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(14)

$\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0)$, the ensemble of values taken by $\varepsilon$ and $\mu$ such as $(\varepsilon + \mu)e(I_n)$ at time $\tau$, knowing that $\mu \in I_n$ at time $\tau$ is independent from the possible values taken by $\varepsilon$ at time 0. Indeed, $\varepsilon$ is a poissonian random process. Moreover, the condition on the $\mu$ value is already fixed at time $\tau$ in $\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0)$, a condition on its value at time 0 does not change the ensemble.

So we can conclude that the ensemble $\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0$ and $\gamma_{\text{emp}}^0$ are independent ensembles, thus:

$$\rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$= \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(15)

and

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0)$$

$$+ \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(16)

For any ensembles $A$ and $B$, one has the basic relationship:

$$P(A | B) = P(B | A)P(A) P(B)$$

so (16) becomes:

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\{(\varepsilon^T + \mu^T)\}e(I_n)) | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(17)

We apply again the law of total probability on the terms $\rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0)$ and $\rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0)$.

$$\rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) = \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

(18)

The same way,

$$\rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) = \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

$$+ \rho((\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \mu_{\text{emp}}^0)$$

(19)

We inject the two last equations in (17):

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0, \gamma_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(20)

$$\rho(\mu_{\text{emp}}^0), \rho(\mu_{\text{emp}}^0)$$ and $\rho(\gamma_{\text{emp}}^0)$ are respectively the probability to have $\mu \in I_n$, $\mu \notin I_n$, and $\{(\varepsilon^T + \mu^T)e(I_n)\}$ at null time. Thus:

$$\rho(\mu_{\text{emp}}^0) = \rho(\mu_{\text{emp}}^0) = \rho(\gamma_{\text{emp}}^0) = \frac{1}{2}$$

and (20) becomes

$$\rho(\gamma_{\text{emp}}^0 | \gamma_{\text{emp}}^0) = \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

$$+ \rho(\gamma_{\text{emp}}^0 | \mu_{\text{emp}}^0) \rho(\mu_{\text{emp}}^0 | \gamma_{\text{emp}}^0)$$

(21)

The probability for the TLS to be in a right spectral window, knowing that it was in it at $t=0$, is the sum of four probabilities, describing the four possible configurations for the occurrence of a coincidence:

- For the first term, the homogeneous line is in the R spectral window at $t=0$ and also at $t=\tau$.

- For the second, the homogeneous line is out of the R spectral window at $t=0$ and in at $t=\tau$. 


we approximate it as a lorentzian centered on homogeneous linewidth is of relative complex shape [11], of) the right spectral window. Despite the fact that the τ window and its total area: the TLS to emit a photon in the right spectral window is be detected. the spectral selection of the right part of the inhomogeneous distribution of the possible µ positions. This distribution is cut by the probability density imposed by the gaussian function.

\[ \rho(\gamma^\tau_{in} | \mu^\tau_{in(out)}) = \frac{\rho(\gamma^\tau_{in} \cap \mu^\tau_{in(out)})}{\rho(\mu^\tau_{in(out)})} = 2\rho(\gamma^\tau_{in} \cap \mu^\tau_{in(out)}) \]

(22)

We consider a Lorentzian centered in the right spectral window (ie. µϵI), and calculate the probability to measure a photon emitted in the right spectral window.

![Diagram](image)

Figure 4. Lorentzian centered on energy µ and gaussian distribution of the possible µ positions. This distribution is cut by the spectral selection of the right part of the inhomogeneous line. It defines the area of the lorentzian where photons can be detected.

For a given lorentzian centered in µ, the probability for the TLS to emit a photon in the right spectral window is the ratio between the lorentzian area in the right spectral window and its total area:

\[ P_\mu(\gamma^\tau_{in}) = \frac{1}{\int_{E_{\mu I}} \text{lor}(\sigma, E - \mu) dE} \int_{E_{\mu I}} \text{lor}(\sigma, E - \mu) dE \]

(23)

with

\[ \text{lor}(\sigma, E - \mu) = \frac{2}{\pi \sigma} \frac{E - \mu}{(E - \mu)^2 + \sigma^2} \]

We have then to consider the probability for the lorentzian to be centered at the energy E between µ and µ + dµ. It is obtained in multiplying the previous expression by the probability density imposed by the gaussian distribution Gauss(Σ, µ) dµ and integrating over all µ such as µϵI.

\[ \rho(\gamma^\tau_{in} \cap \mu^\tau_{in(out)}) = \int_{\mu_{in(out)}}^{\mu_{out}} \text{Gauss}(\Sigma, \mu) \int_{E_{\mu I}} \text{lor}(\sigma, E - \mu) dE d\mu \]

(24)

with Gauss(Σ, µ) = \( \frac{1}{\Sigma \sqrt{2\pi}} \exp \left( -\frac{\alpha^2}{2\Sigma^2} \right) \), \( \Sigma_s = \frac{\Sigma}{2\sqrt{2\ln 2}} \)

the standard deviation and Σ the full width at half maximum of the gaussian function.

This is the probability for the TLS to be in the right spectral window and the lorentzian centered in the right (left) spectral window at any time, so it does not depend on the delay τ and are only functions of the homogeneous linewidth σ and the gaussian standard deviation ΣS:

\[ \rho(\gamma^\tau_{in} \cap \mu^\tau_{in(out)}) = \rho(\gamma^\tau_{in} \cap \mu^\tau_{in(out)}) = \alpha_{in(out)}(\sigma, \Sigma) \]

(25)

We rewrite the eq. 21 with these coefficients:

\[ \rho(\gamma^\tau_{in} | \mu^\tau_{in}) = 4[\alpha^2_{in}\rho(\mu^\tau_{in} | \mu^\tau_{in}) + \alpha_{in}\alpha_{out}\rho(\mu^\tau_{in} | \mu^\tau_{out}) + \alpha_{out}\alpha_{in}\rho(\mu^\tau_{out} | \mu^\tau_{in}) + \alpha^2_{out}\rho(\mu^\tau_{out} | \mu^\tau_{out})] \]

(26)

This probability depends on one hand on the coefficients α_{in} and α_{out} which depend themselves on the linewidth of the homogeneous lorentzian line and of the gaussian distribution. They take into account the random uncorrelated statistics introduced by the finite homogeneous linewidth. On the other hand, eq. 26 depends on the probabilities \( \rho(\mu^\tau_{in} | \mu^\tau_{in}) \), \( \rho(\mu^\tau_{in} | \mu^\tau_{out}) \), \( \rho(\mu^\tau_{out} | \mu^\tau_{in}) \), \( \rho(\mu^\tau_{out} | \mu^\tau_{out}) \) which are describing how the mean value of the homogeneous linewidth is transiting from one spectral window to the other and take into account the time-correlated part of the statistics. All these probabilities can be determined analytically with the model of the infinitely sharp homogeneous linewidth presented in the reference [9], and its associated rate equations.

Because of the configuration chosen in this letter (ie. left side versus right side of the spectrum), the probability γ_{R} for the mean value of the homogeneous line to jump from the right spectral window to the left is the same as the probability γ_{L} for the homogeneous line to make the opposite move, thus:

\[ \gamma_R = \gamma_L = \frac{\gamma_c}{2} \]

(27)
Then, we can derive:
\[
\rho(\mu_{in}^r | \mu_{out}^0) = \frac{\gamma R}{\gamma c} + (1-\frac{\gamma R}{\gamma c})\exp(-\gamma_c \tau) = \frac{1}{2} \left(1 + \exp(-\gamma_c \tau)\right)
\]  \tag{28}
\[
\rho(\mu_{in}^r | \mu_{out}^0) = \frac{\gamma R}{\gamma c} [1 - \exp(-\gamma_c \tau)]
\]  
\[
= \frac{1}{2} [1 - \exp(-\gamma_c \tau)]
\]
\[
\rho(\mu_{out}^0 | \mu_{in}^r) = \frac{\gamma R}{\gamma c} [1 - \exp(-\gamma_c \tau)]
\]  
\[
= \frac{1}{2} [1 - \exp(-\gamma_c \tau)]
\]
\[
\rho(\mu_{out}^0 | \mu_{out}^0) = \frac{1}{2} [1 + \exp(-\gamma_c \tau)]
\]  \tag{29}

We put all these expressions in eq. 26:
\[
\rho(\gamma_{in}^r | \gamma_{in}^0) = 4 \frac{1}{2} (\alpha_{in}^2 + \alpha_{out}^2) + \alpha_{in}\alpha_{out}
\]
\[
+ \left(\frac{1}{2} (\alpha_{in}^2 + \alpha_{out}^2) - \alpha_{in}\alpha_{out}\right) \exp(-\gamma_c \tau) \tag{30}
\]

To obtain the correlation function, we divide this expression by \(\rho(\gamma_{in}^\infty | \gamma_{in}^0) = 4 \left(\frac{1}{2} (\alpha_{in}^2 + \alpha_{out}^2) + \alpha_{in}\alpha_{out}\right)\),
\[
g_{in}^2(\tau) = 1 + \left(\frac{1}{2} (\alpha_{in}^2 + \alpha_{out}^2) - \alpha_{in}\alpha_{out}\right) \exp(-\gamma_c \tau) \tag{31}
\]
finally, rearranging terms and multiplying the emission part \(g_{in}^2(\tau)\), one has:
\[
g^2(R, R, \tau) = [1 + \frac{(\alpha_{in} - \alpha_{out})^2}{(\alpha_{in} + \alpha_{out})^2} \exp(-\gamma_c \tau)]
\]
\[
\times \left[1 - \exp(-(r + \gamma \tau))\right] \tag{32}
\]

Thus, the HLAF is on the form:
\[
g^2(R, R, \tau) = [1 + \beta.\exp(-\gamma_c \tau)][1 - \exp(-(r + \gamma \tau))] \tag{33}
\]
with,
\[
\beta = \frac{(\alpha_{in} - \alpha_{out})^2}{(\alpha_{in} + \alpha_{out})^2} \tag{34}
\]

We can make two remarks about this factor:
- The bunching factor \(\beta\) is exclusively ruled by the uncorrelated random statistics of the finite homogeneous linewidth (expressed by coefficients \(\alpha_{in}\) and \(\alpha_{out}\)), and is a function of \(\sigma\) and \(\Sigma\). As shown in fig. 5, the bunching is important (red color) for small \(\sigma\) and large fluctuation amplitudes \(\Sigma\).
- The behavior of \(\beta\) at the limits is interesting: when the homogeneous linewidth tends to 0, i.e. \(\sigma \ll \Sigma\), we have:
\[
\alpha_{in} \to \frac{1}{2}
\]
\[
\alpha_{out} \to 0
\]
so, \(\beta \to 1\), which is its expected value in the case of the infinitely sharp linewidth.

When the homogeneous linewidth becomes larger than the standard deviation of the fluctuation, \(\sigma \gg \Sigma\):
\[
\alpha_{in} \to \frac{1}{4}
\]
\[
\alpha_{out} \to \frac{1}{4}
\]
and \(\beta \to 0\), the bunching is collapsing. This situation corresponds to a case in which the poissonian statistics of the homogeneous linewidth takes over the correlated statistics of the spectral diffusion.

The HLAF bears simultaneously the signature of the subpoissonian emission statistics (zero delay dip), the correlated spectral diffusion energy statistics (bunching), and the poissonian energy statistics of the homogeneous linewidth (degradation of the bunching), the two last signatures having opposing effects.

\section*{V. MONTE-CARLO SIMULATION}

In order to confirm the analytical expression derived from the calculations above, we performed a Monte Carlo simulation, by building numerically the function \(g^2(R, R, \tau)\) for an emitter spectrally diffusing in and out of the detection spectral window. We do not take into account the subpoissonian statistics of the emitted since it can be factorized in the correlation function (see eq. 33). Thus, the simulation gives a direct access to \(g_{in}^2(\tau)\) and consequently to \(\beta\) since
\[
\beta = g_{in}^2(0) \tag{35}
\]

After generating a poissonian stream of photons, we assign to each of them a random energy in a gaussian distribution. We calculate the probability of the mean value of the homogeneous line to jump between the emission for the \((i-1)th\) photon and the \(i^{th}\) photon.
The energy of the \((i-1)\)th photon is in the interval:

\[
I_{i-1} = I_{in} = \left[ E_{i-1} - \frac{\delta E}{2}, E_{i-1} + \frac{\delta E}{2} \right]
\]

with \(\delta E\) an infinitesimal energy.

In the case of a jump of the homogeneous line, the energy of the \(i\)th photon is in the interval:

\[
I_i = I_{out} = \left[ -\infty, E_{i-1} - \frac{\delta E}{2} \right] \cup \left[ E_{i-1} + \frac{\delta E}{2}, +\infty \right]
\]

Thus, the probability for the homogeneous line to jump between the emission of the \(i\)th and the \((i-1)\)th photon is:

\[
P_{\text{jump}(i)} = \rho \{ \mu^e I_{out} \mid \{ \mu^0 e I_{in} \} \} = \frac{\gamma_{out}}{\gamma_c} \text{exp}(-\gamma_c \tau)
\]

This probability has already been calculated in the previous section (see equation (23)).

\(\gamma_{out}\) is the exit rate of the homogeneous line, from \(I_{in}\) to \(I_{out}\), \(\gamma_c\) is the “jump rate” of the homogeneous line.

For an infinitely small \(\delta E\), \(\gamma_c \approx \gamma_{out}\) and:

\[
P_{\text{jump}(i/(i-1))} = [1 - \text{exp}(-\gamma_c \tau)]
\]

If the \(i\)th photon does not jump, it takes the same energy than the \((i-1)\)th photon.

We then discriminate the photons belonging to energies outside the detection area by applying an energy condition. They will not be counted in the correlation process.

To calculate the correlation function of the resulting photons stream, we compute the delay between the arrival of each photon and all the other photons of the stream. By building the histogram of these delays, one obtains the correlation function of the stream [15].

The result of the simulation is plotted on fig. 6 a). We find again the result explained in the previous section, the bunching part of the HLAF takes a value of 2 at zero delay. To evaluate \(b\) from the simulated data, one only need to extract \(g^2(0)\).

To simulate the finite homogeneous linewidth effect, we add for each photon an energy shift \(\varepsilon\). \(\varepsilon\) is a random variable distributed along a lorentzian distribution of linewidth \(\sigma\). The effect of this addition is shown on fig. 6 b). As expected the bunching is less marked and the \(b\) factor goes from 1 to 0.5 in the case of a finite homogeneous linewidth of \(\sigma = 0.3 \text{meV}\) and a fluctuation amplitude of \(\Sigma = 1.7 \text{meV}\).

To compare this simulation with the analytical expression, we fix the \(\Sigma\) parameter and change \(\sigma\). We then report the values of the \(b\) factor versus the ratio \(\frac{\Sigma}{\sigma}\) in both cases (see fig. 7). We can notice that the two methods (analytical and Monte Carlo) give the same dependence.

\[
S(\sigma, \Sigma, \omega) = \int_{-\infty}^{+\infty} \text{lor} \left( \sigma, \xi \right) \text{gauss} \left( \Sigma, \xi - \omega \right) d\xi
\]

Figure 6. Calculated half line autocorrelation functions of a poissonnian emitter spectrally diffusing. a) for an infinitely small homogeneous linewidth, b) for a homogeneous linewidth \(\sigma = 0.3 \text{meV}\).

Figure 7. \(b\) factor versus the ratio between homogeneous linewidth \(\sigma\) and fluctuation amplitude \(\Sigma\) for fixed value of \(\Sigma\). Red line: analytical model described by eq. Blue dots: Monte Carlo simulation result.

VI. HOMOGENEOUS AND INHOMOGENEOUS LINewidth DETERMINATION

The fit of the experimental data allows to extract, as two independent parameters, the correlation time \(\tau_c\) and \(b\). As shown in the previous section the latter depends on the fluctuation amplitude \(\Sigma\) and the homogeneous linewidth \(\sigma\). To estimate them separately, we can use the emission spectrum measured, and by making the same assumptions as before (the homogeneous line is lorentzian and the fluctuation distribution is gaussian) we obtain a second equation linking these two parameters since, in this case, the emission spectrum has a Voigt profile:

\[
S(\sigma, \Sigma, \omega) = \int_{-\infty}^{+\infty} \text{lor} \left( \sigma, \xi \right) \text{gauss} \left( \Sigma, \xi - \omega \right) d\xi
\]
VII. CONCLUSION

The HLAF is not only a function of the correlation time of the spectral diffusion. It is dependent on the nature of the photon energy statistics. From the calculation we performed, one can interpret analytically the importance of these different statistics in the observed correlation function. By measuring the HLAF and by making a temperature dependence of a spectrally diffusing emitter it is possible to describe how electronic fluctuations and phonon broadening are evolving, and which one is dominating the spectrum. Thus, PCS technique can bring all the informations for the full characterization of a spectrally diffusing emitter. Indeed, after data treatment technique presented in the last section of this letter, one can obtain separately $\sigma$ (homogeneous linewidth), $\Sigma$ (fluctuation amplitude) and $\gamma_c$ (correlation rate). Other existing methods [16, 17], except the photon correlation Fourier spectroscopy method (PCFS) [18, 19] are not bringing all these informations at the same time and all of them are not adapted in the case of fast fluctuations. From an experimentalist point of view, it is important to note that the spectral resolution of the PCS experiment is limited by the spectrometer, which makes it less accurate than PCFS, which takes advantage of the Fourier transform spectroscopy. However the PCS technique is extremely easy to setup and does not require the drastic stability conditions demanded by the PCFS technique. Since photons correlation experiments require long integration time, such sensitivity led to the degradation of the time resolution to 20 $\mu$s for the PCFS technique, which forbids to probe fast fluctuations of the energy emitter. In PCS technique, time resolution is only limited by photodiodes and can be lowered to around 100 ps. This technique is a reliable and accessible technique for a complete and fast characterization of a single photon emitter in a solid state environment.

[1] R. Hanbury Brown & R. Q. Twiss, Nature 177, 27 - 29 (1956)
[2] H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. 39, 691–695 (1977)
[3] H. J. Kimble, and L. Mandel, Phys. Rev. A 13, 2123–2144 (1976)
[4] E. Knill, R. Laflamme & G. J. Milburn, Nature 409, 46 (2001)
[5] C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. 59, 2044–2046 (1987)
[6] R. G. Neuhauser, K. T. Shimizu, W. K. Woo, S. A. Empedocles, and M. G. Bawendi Phys. Rev. Lett. 85, 3301 (2000)
[7] K. Wodkiewicz, B. W. Shore, and J. H. Eberly, Phys. Rev. A, 30(5) :2390, 1984.
[8] K. Wodkiewicz, B. W. Shore, and J. H. Eberly, J. Opt. Soc. Am. B, 1(3) :398, 1984.
[9] G. Sallen, A. Tribu, T. Aichele, R. André, L. Besombes, C. Bougerol, M. Richard, S. Tatarenko, K. Kheng and J. Ph. Poizat, Nature Photon. 4, 696 (2010)
[10] Mandel and Wolf. Optical coherence and quantum optics, chapter 14, sec 2.
[11] L. Besombes, K. Kheng, L. Marsal, and H. Mariette, Phys. Rev. B 63, 155307 (2001)
[12] R. Kubo. Note on the Stochastic theory of resonance Absorption. J. Phys. Soc. Jap, 9(6) :935, 1954.
[13] P. W. Anderson, J. Phys. Soc. Jpn., 9 :316, 1954.
[14] G. Sallen, A. Tribu, T. Aichele, R. Andre, L. Besombes, C. Bougerol, M. Richard, S. Tatarenko,K. Kheng and J.-Ph. Poizat, Supplementary informations Nature Photonics (2010)
[15] D. Meiser and M. J. Holland, Phys Rev A 81, 063827 (2010).
[16] Phedon Palinginis, Sasha Tavenner, Mark Lonerga, and Hailin Wang, Phys. Rev. B 67, 201307(R) (2003)
[17] Taras Plakhotnik and Daniel Walser, Phys. Rev. Lett. 80, 4064-4067 (1998)
[18] L. Coolen, X. Brokmann, P. Spinicelli, Phys Rev Lett., 100, 027403, (2003)
[19] L. Coolen, X. Brokmann, P. Spinicelli, and J.-P. Hermier Phys. Rev. Lett. 100, 027403 (2008).