9.2 Photon arrival trajectories

We unraveled the master equations into elementary photon detection events following the formal structure of quantum measurements in a meaningfully short but finite detection time. Our next destination is to interpret the photodetection record, i.e. the *photon arrival trajectory*, as a sequence of elementary dichotomous measurements and its meaningful integration into a continuous-time picture.

An individual photon arrival trajectory is irreproducible like any other quantum measurement. Yet, the correlations between arrival times can be estimated along the trajectories and compared to predictions founded by the ergodic hypothesis establishing the correspondence between time and ensemble averages. Eq. (220) is a nice motivational example of what kind of theory can be expected: it connects two emission events (234) by a propagator derived from the Bloch equations. The detail of algorithm depends on the protocol used for photon counting, here we distinguish two cases (numerics compared at Fig 7).

- The detector is checked for the entire experiment. To calculate the probability of a record we partition the time window between 0 and T to many short intervals Δt, in which a photon was detected (y) or not (n). The restricted density matrix for each sequence of outcomes is constructed following the rules of Eqs (234) and (235). For instance, the sequence y, n, n, y, n, n, n, y has probability

$$P(y,n,n,y,n,n,n,y) = \mathrm{Tr}\, \breve{R}\Delta t \left\{\breve{1} + \left(\breve{\mathcal{L}} - \breve{R}\right)\Delta t\right\}^3 \breve{R}\Delta t \times \left\{\breve{1} + \left(\breve{\mathcal{L}} - \breve{R}\right)\Delta t\right\}^2 \breve{R}\Delta t \hat{\rho}(0) \qquad (236)$$

  Since we can choose $\Delta t \ll 1/\Gamma$, there is a large number m of photonless intervals between two consecutive photon detections

$$\left\{\breve{1} + \left(\breve{\mathcal{L}} - \breve{R}\right)\Delta t\right\}^m \to \mathrm{e}^{(\breve{\mathcal{L}} - \breve{R})m\Delta t}$$

  so this combined propagator only depends on their delay. We can now switch to a general trajectory described by the arrival times $\tau_1, \tau_2, \ldots \tau_N$. Its probability on the time window $(0, T)$ is recast as

$$\frac{P(\tau_1, \tau_2, \ldots, \tau_N)}{(\Delta t)^N} = \mathrm{Tr}\, \mathrm{e}^{(\breve{\mathcal{L}} - \breve{R})(T-\tau_N)} \breve{R} \ldots \breve{R} \mathrm{e}^{(\breve{\mathcal{L}} - \breve{R})(\tau_2-\tau_1)} \breve{R} \mathrm{e}^{(\breve{\mathcal{L}} - \breve{R})\tau_1} \hat{\rho}(0).$$

The initial density matrix $\hat{\rho}(0)$ is set to the steady state (Eq. (222)) defined by $\breve{\mathcal{L}}\hat{\rho} = 0$. The propagator $\mathrm{e}^{(\breve{\mathcal{L}}-\breve{R})t}$ is solution to equation

$$\frac{\mathrm{d}\hat{\rho}}{\mathrm{d}t} = (\breve{\mathcal{L}} - \breve{R})\hat{\rho} \tag{237}$$

and describes evolution without emission event (solid line at Fig 7). Eq. (237) does not preserve total population, $\mathrm{Tr}\hat{\rho}(t)$ decays, and time elapsed between the start (end) and the first (last) emission matters. Next, this protocol is sensitive to incomplete detection. The realistic detector only detects some part p of emitted photons and can be analyzed by modifying the resetting operator $\breve{R} \to p\breve{R}$. The recalculation is not difficult, but the probability for photon arrival trajectory does not scale with p in any simple way.

- We check the detector for a photon in an interval Δt around time 0 and again after delay T. The detector is not checked inside the window $(0, T)$, any number of photons can be emitted during it. Evolution between the detection intervals is thus calculated by solving the standard RWA Bloch Eq. (219), i.e. the propagator is the (trace-preserving) evolution operator $\mathrm{e}^{\breve{\mathcal{L}}T}$ and this protocol corresponds to solutions given above by Eqs. (220) or (221). The steady state is preserved $\mathrm{e}^{\breve{\mathcal{L}}t}\hat{\rho}_{\mathrm{s}} = \hat{\rho}_{\mathrm{s}}$, so the first detection at initial time 0 was no special. The probability of the joint detection is given by

$$\frac{P(y, y; 0, T)}{(\Delta t)^2} = \mathrm{Tr}\ \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho}_{\mathrm{s}}. \tag{238}$$

Represented at Fig 7 by dashed lines, it is also called fluorescence autocorrelation, i.e. the correlation function $\langle I(T)I(0)\rangle$ of the fluorescence intensities I. Similarly the joint probability density for N detections attempted at times $\tau_1, \tau_2, \ldots, \tau_N$ is calculated using

$$\frac{P(y, y, \ldots; \tau_1, \tau_2, \ldots, \tau_N)}{(\Delta t)^N} = \mathrm{Tr}\ \breve{R}\mathrm{e}^{\breve{\mathcal{L}}(\tau_N - \tau_{N-1})}\breve{R}\ \ldots \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T(\tau_2 - \tau_1)}\breve{R}\hat{\rho}_{\mathrm{s}}.$$

Important advantage of this protocol is a power law p^N scaling for the incomplete detection. Normalized autocorrelation

$$g(T) \equiv \frac{\langle I(T)I(0)\rangle}{\langle I\rangle^2} = \frac{P(y, y; 0, T)}{P(y, 0)P(y, T)}$$

is therefore independent of p.

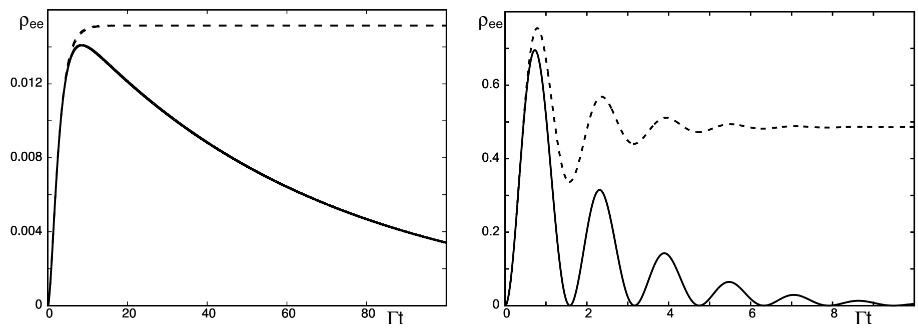

Figure 7: Probability for the second photon arrival vs. fluorescence autocorrelation. Left panel: Overdamped solution for autocorrelation in weak fields $\mathcal{E}/\Gamma = 0.125$ (dashed, Eq. (220)) starts with the dark period after resetting and assumes a stable asymptotic value as emissions are decorrelated at longer delays. The second photon arrival, after the initial dark time, is following slow decay of the probability that no photon has been recorder yet (Eq. (237), solid). Right: Rabi oscillations of strong fields $\mathcal{E}/\Gamma = 4.0$ shown in the regular dark periods of the second photon arrivals, are gradually desynchronized by resetting at random emission times in autocorrelation (221).

The heavy use of conditional probabilities, freedom to choose a protocol, differences between associated propagators is reminiscent of a type of probabilistic estimates of knowledge (belief) called Bayesian statistics.

In conclusion we note, that the outlined algorithm can be easily extended for broadband detection beyond the single two-level molecule by proper reinterpretation of resetting matrix, which must sum over all relevant transitions. For instance, emission from two independent (e.g. identical) two-level molecules is associated with the following resetting

$$\hat{\rho}_y^{(1,2)} = \breve{R}\hat{\rho}^{(1)} \otimes \hat{\rho}^{(2)} + \hat{\rho}^{(1)} \otimes \breve{R}\hat{\rho}^{(2)}$$

Autocorrelation $P^{(1,2)}$ can be factorized into the single-molecular P

$$\begin{aligned}
\frac{P^{(1,2)}(y,y;0,T)}{(\Delta t)^2} &= \mathrm{Tr}\ \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho}^{(1)} \otimes \mathrm{e}^{\breve{\mathcal{L}}T}\hat{\rho}^{(2)} + \mathrm{Tr}\ \mathrm{e}^{\breve{\mathcal{L}}T}\hat{\rho}^{(1)} \otimes \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho}^{(2)} \\
&+\mathrm{Tr}\ \mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho}^{(1)} \otimes \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\hat{\rho}^{(2)} + \mathrm{Tr}\ \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\hat{\rho}^{(1)} \otimes \mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho}^{(2)} \\
&= 2\mathrm{Tr}\ \breve{R}\mathrm{e}^{\breve{\mathcal{L}}T}\breve{R}\hat{\rho} + 2\left(\mathrm{Tr}\ \breve{R}\hat{\rho}\right)^2 = 2\frac{P(y,y;0,T)}{(\Delta t)^2} + 2I^2
\end{aligned}$$

The normalized difference against random emission

$$\frac{P^{(1,2)}(y,y;0,T) - (2I)^2(\Delta t)^2}{(2I)^2(\Delta t)^2} = \frac{1}{M}\frac{P(y,y;0,T) - I^2(\Delta t)^2}{I^2(\Delta t)^2}$$

is reduced by $M = 2$, the number of molecules.