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## Chapter 9

# Coherence and statistical properties of ultra-weak photon emission

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**Abstract:** We present a critical review of the works related to the statistical properties of biological ultra-weak photon emission (UPE) and in particular to its coherence properties. Starting with a brief description of the concept of coherence in quantum and classical physics, we then review models used to analyze photon distributions obtained from measurements of UPE. Moreover, we review experiments focused on statistical properties of UPE and try to assess them from the point of view of current understanding of physics and biophysics. A critical study of the results and their interpretations leads us to conclude that there is no undoubted proof of the coherence of UPE. We highlight particular problems of past research with respect to data interpretation or hypothesis building when looking for coherent light sources or emission and discuss the application of standard quantum optical methods for assessing the coherence of an optical field. Since emerging studies show that not only coherence properties but also fractal and chaotic properties of UPE time series signals can be analyzed, we outline briefly these fractal and chaotic properties of UPE time series presenting them as a possible new avenue for UPE signal analysis.

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## 1. Introduction

From the very beginning (1920s) of biological ultra-weak photon emission (UPE) research, scientists were wondering whether the light spontaneously emitted by biological cells exhibits any exceptional signal property. Especially with the arrival of quantum optical theory of coherence (1960s), leading for example to the development of lasers, it became obvious that light can manifest very special properties with coherence being the most remarkable one. It was hypothesized for biological systems that their internal electromagnetic field is a coherent field and, further, that this coherence plays a significant role in pattern formation of biological systems (Pokorný and Wu, 1998; Popp, 2005; Cifra, 2012), since it is well known in physics that coherent electromagnetic fields can interfere and form stable space-time patterns.

Furthermore, research of statistical properties of UPE signals (not limited to coherence) is intriguing because it does not deal only with quantity (intensity) and color (energy) of the detected light but also with its quality (orderliness, informational content). Such research is basically focused on quantifying the time sequences of photons released from an optical field using various physical and mathematical methods (e.g., for quantifying photocount distribution, or for assessing fractal and chaotic properties). In addition to obtaining a new feature for getting “fingerprint” photons possibly leading to applications in diagnostics, these statistical properties of UPE signals bring also insight into the physical nature of light from biological systems and moreover, the photon generating processes therein.

In the following we present (i) a brief description of the concept of coherence for so-called quantum and classical cases. We then review (ii) models used to analyze distributions of photons emitted from biological systems. We also review (iii) experiments focused on statistical properties of UPE and try to assess them from the point of view of current understanding of physics and biophysics. Finally, we outline (iv) fractal and chaotic properties of UPE time series as a possible new avenue for UPE signal analysis.

## 2. Coherence of light

Coherence is one of the fundamental statistical properties of light and yet quite subtle. In a nutshell, coherence is the ability of light to build interference. This is (according to Grimaldi) the fact that darkness can be obtained by adding light to light<sup>1</sup> (Grimaldi, 1665, p. 189). Broadly speaking, light beams are coherent if they combine like waves (by adding the amplitudes of the beams) while they are incoherent if they combine like particles (by add-

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<sup>1</sup> *obscuratio, facta per solam additionem luminis*. In fact, Grimaldi did not really observe interference (Kipnis, 1991, p. 135), but his happy turn of phrase was remembered.

ing the intensities, *i.e.* the square of the amplitudes, of the beams).

As a consequence of these subtle effects, which light displays dependent on the experimental setup, statements about coherence demand and in fact are supported by solid proofs presented in the scientific literature. In contrast, many of the papers from the period 1980–2005 on coherence of ultra-weak photon emission contain speculative statements inspired (or not) by experimental results. Hence, the important purpose of this chapter is to assess, in the context of currently accepted viewpoints in physics, the solidity of the conclusions that the authors have drawn from their data.

Some basic terminological relations should be explained in the beginning. The terminology used in quantum mechanics and quantum field theory often occurs in UPE literature, where the term coherence may refer either to (i) wave functions solving the Schrödinger equation or (ii) light, which is, strictly speaking, not the same (although related) and often creates confusion. In quantum mechanics, coherence is an intrinsic property of wave functions and once decoherence occurs (*i.e.* loss of wave function coherence – collapse of wave function), the system often behaves classically. Therefore, quantum behavior is equated to coherence by some authors, but it is reasonable only when speaking about wave functions.

In this chapter, which deals with the coherence of light, one cannot directly equate either *non-classical (quantum)* with *coherent* light, or *classical* with *incoherent* light. Generally, the quantum optical framework can explain all states of light. The classical framework can explain only some of them and those can be called classical. The states which can be only explained in a quantum framework are usually called purely quantum states. The coherence of light can be both of classical and quantum character, thermal states of light (see below) can be described in classical and quantum framework, while certain states can be described only in a quantum framework (*e.g.* some squeezed states).

## 2.1. Classical vs. quantum coherence of light

The coherence in classical physics typically describes how the intensities of two waves combine. If the intensity of the combined wave is the intensity of the sum of the amplitudes of the two waves, then we say that the waves are fully coherent and interference effects are maximal<sup>2</sup>. If the intensity of the combined wave is the sum of the intensities of the two waves, then we say that the waves are fully incoherent and do not interfere. Partial coherence describes intermediate situations between incoherent and fully coherent waves. Note that there are several types of coherence, which influence the visibility of interference fringes: temporal (refers to the correlation of the field between two times), spatial (refers to the correlation between two space points), spectral (refers to the correlation of field frequencies between

two points) and polarization (refers to the correlation of the polarization of light fields).

A quantum field theory of optical coherence is given by Mandel and Wolf (see Chapter 12 in Mandel and Wolf, 1995). Mathematically, instead of describing the light field by functions as in the classical case, a quantum treatment describes the light field by *operators* and *quantum states*. Field operators destroy (annihilation operator) or create (creation operator) a “particle” of the field – a photon. A major consequence from quantum optics is that light can behave in ways that are inexplicable by classical physics, *e.g.* noise reduction in squeezed states, or entanglement of photons over a large distance.

### 3. Models for UPE photons and their photocount distributions

The coherence properties of UPE were investigated experimentally mainly by measuring the distribution of counts produced by UPE photons with a photomultiplier<sup>2</sup>. A few studies were also performed using a CCD camera (*e.g.*, Kobayashi et al., 1999). Before introducing the basic models of states of light for photocount distributions we want to bring awareness of two important aspects.

First, photocount distributions show the probability for a number of counts to be detected in finite time interval  $t$  (see *e.g.* Fig. 1). Such a distribution is one of the tools to describe statistical and coherence properties of light. The motivation behind this method is to relate the photocount distribution to the state of the biological system. Even though photocount distributions are rather easy to measure and give a hint on the possible states of light and do not require any sophisticated measurement system, they cannot unambiguously determine whether UPE is coherent or not. The reason is that specific photocount distributions cannot be uniquely attributed to specific states of light, *i.e.* similar or identical photocount distributions may come from different states of light. Yet, a solid method that quantifies coherence time or length of radiation comes from interferometric measurements. But while they are standard in quantum optics, they were not so far applied to UPE from biological systems, mainly because of experimental difficulties, which arise from the small number of photons and the nonstationarity of biological systems. Besides, the quantum nature of UPE was already suggested by Walter Stempell in 1932 (long before quantum electro-

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<sup>2</sup> Photon detectors do not measure the amplitude of light (*i.e.* its electric field  $E(\mathbf{r}, t)$ ) but its intensity, which is the average over time of the square of the amplitude of light.

dynamics was fully established)<sup>3</sup>. Indeed, since chemiluminescence is ultimately interpreted as a quantum phenomenon (photons are quantum objects), the source of UPE is quantum mechanical in nature. However, when a photocount statistics is considered to be classical, it is not because its source is classical but because it can be described by a (positive) probability distribution.

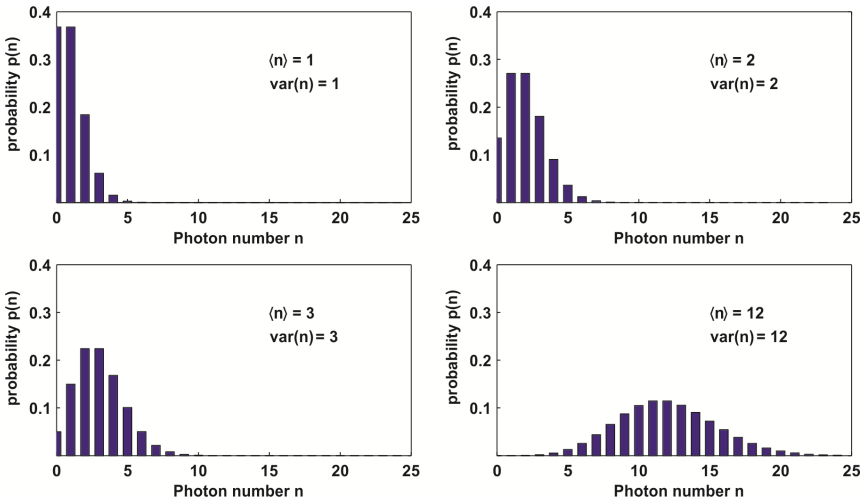
In the literature on UPE, the most commonly encountered types of states of light are the so-called coherent, squeezed, and thermal states, which we explain in the following section.

Coherent states were discovered by Schrödinger (1926), rediscovered by Schwinger (1953), and further studied by Glauber (1963) who called them coherent states. Coherent states are now a standard tool of quantum optics (Mandel and Wolf, 1995). From the conceptual point of view, coherent states are those quantum states that correspond to classical electromagnetic waves. For instance, a classical varying current (a simple source of electromagnetic waves – a piece of electric wire carrying a macroscopic varying current,  $I(t)$ , for instance) gives rise to a coherent state of the photon field (Itzykson and Zuber, 1980). The photocount statistics of a system in a coherent state gives rise to a Poisson distribution (see Fig. 1)<sup>4</sup>. A Poisson distribution is a sign of classical light field. Its variance is equal to its mean:  $\langle \Delta n^2 \rangle = \langle n \rangle$  (see also Fig. 1). Departure from a Poisson distribution can be measured by the so-called Fano factor  $F$  such that  $F = \langle \Delta n^2 \rangle / \langle n \rangle$  or by the Mandel parameter  $Q = F - 1$ . A photocount statistics is super-Poissonian if  $F > 1$  and  $Q > 0$ , it is sub-Poissonian if  $F < 1$  and  $Q < 0$ . A super-Poissonian distribution can be classical but a sub-Poissonian distribution is a purely quantum state of light: it cannot be described by a positive probability distribution.

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<sup>3</sup> According to Stempell, “... die Quantennatur der Strahlung dabei in die Erscheinung tritt.” (Stempell, 1932, p. 63).

<sup>4</sup> Keep in mind that also other states of light can yield Poisson-like distribution of photocount statistics.



**Figure 1.** Poisson distributions for four different average values of photon counts  $\langle n \rangle$ . A reading example: At an average signal intensity of  $\langle n \rangle = 3$  counts per time interval  $t$ , the probability to detect 5 counts in time interval  $t$  is 0.1 (*i.e.* 10%).

However, one has to be careful to avoid experimental and instrumental artifacts, which can also lead to observations different from Poisson distribution (*e.g.* super- or sub-Poisson) of photocounts even when measuring classical and thermal light that would normally lead to a Poissonian distribution if it was measured correctly. Super-Poisson distribution can be caused by nonstationarity of the light source such as (i) a modulation intensity of the photon signal by periodic internal or external factors or (ii) bursts of photon emission caused by stochastic processes. However, super-Poisson distribution caused by such nonstationarities has nothing to do with squeezed states of light.

In squeezed states, the dispersion (uncertainty) of one variable is reduced at the cost of an increase in the dispersion of the other canonical variable (amplitude vs. phase, or position vs. momentum). Various squeezed states were used in the UPE literature, but the most general ones are called two-photon coherent states (generalization of states which have minimum uncertainty) (Yuen, 1976). They have become standard states of quantum optics (Mandel and Wolf, 1995, p. 1046) and their photocount statistics is known (Mandel and Wolf, 1995, p. 1050). Squeezed states are interesting because they can manifest lower intrinsic noise (fluctuations around mean) than coherent light, a feature which classical light cannot achieve (see also

Fig. 2). The lower the intrinsic noise (related to uncertainty), the higher the efficiency of such states to transmit information.

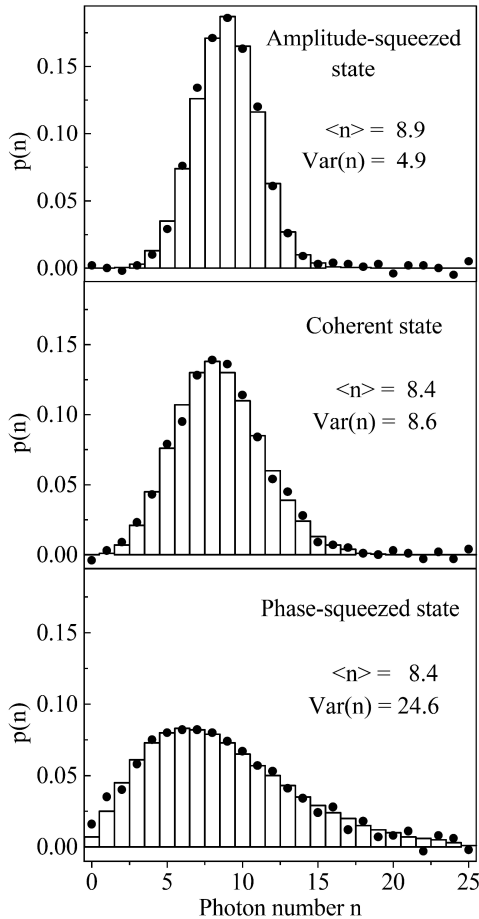
A thermal state of light can be obtained by filtering a spectral band from a blackbody (thermal) radiation. Thermal states are classical, *i.e.* they can be described within the framework of classical physics, and represent a model of random light with very low coherence. Usually a thermal light is not emitted by a single oscillator but by a large number of oscillators (also called modes or degrees of freedom). The number of degrees of freedom  $M$  can be estimated as the product of a time degeneracy ( $M_t$ ) and a space degeneracy ( $M_s$ ), where the time degeneracy is the ratio of the measurement interval  $t$  over the coherence time (Loudon, 2000, p. 97) and the space degeneracy is the number of incoherent oscillators in the source (Mosset, 2004). The photocount statistics of a thermal source depends on the number of degrees of freedom  $M$  (Mandel and Wolf, 1995, p. 680 and 731) (see also Fig. 3).

Since the question whether photons are in a coherent or a thermal state is recurrent in the UPE literature, it is important to know how to distinguish between them. However, since photocount statistics of thermal light becomes equal to that of a coherent state when  $M$  is large, photocount statistics is not able to discriminate between a coherent and a thermal state with many modes.

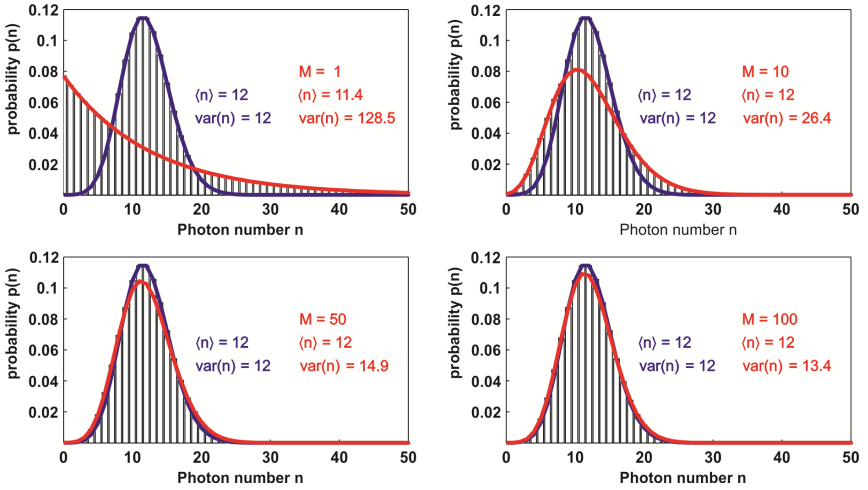
This can in particular be seen by the relation between variance and mean in a thermal state:

$$\langle \Delta n^2 \rangle = \langle n \rangle + \langle n \rangle / M .$$

We see that, when  $M$  is very large, we recover  $\langle \Delta n^2 \rangle = \langle n \rangle$  as for a coherent state (Fig. 3). We know that the number of modes  $M$  is generally very large for chaotic sources (Jiang et al., 2003), bringing the relation between variance and mean of photocount distribution close to that of a coherent state. Therefore, since the average number of photons  $\langle n \rangle$  is particularly small in UPE experiments, we expect that it will be difficult to distinguish thermal UPE from coherent UPE based on photocount distributions.



**Figure 2.** Squeezed state photocount distributions. The upper graph shows a so-called amplitude squeezed light, where fluctuations of the number of photon counts (*i.e.* the amplitude) are reduced leading to a narrower photon distribution. The middle graph shows a Poisson distribution of photons compatible with a coherent light. The lower graph describes phase-squeezed light with reduced phase fluctuations and increased amplitude fluctuations leading to a broader distribution. Dots in all three graphs are measured, *i.e.* observed values from technical tunable squeezed light generating sources, the bars are theoretically calculated, *i.e.* expected values. The graphs are from Breitenbach (1998).



**Figure 3.** The thermal field photocount distribution (blue line) approaches the Poisson distribution (red line) for a large number of modes  $M$ , *i.e.* for a large number of independently radiating sources (molecules, atoms) or from a single source with very short coherence time compared to time interval of measurement. The average intensity of the photon signal  $\langle n \rangle$  is the same for all displayed distributions.

#### 4. Experimental measurements of the photocount statistics of UPE

There are several tens of experimental works which aimed to study photocount statistics of biological UPE related sources from different species from bacteria to man. List of them can be found in Table 1. We assess the works which characterize the state of art most accurately in this section.

| Sample from                     |                                   | References  |
|---------------------------------|-----------------------------------|---|
| <b>Chemicals</b>                |                                   |   |
| Luminol                         | $C_8 H_7 N_3 O_2$                 | Shen et al., 1993   |
| Polystyrol                      | $(C_8 H_8)_n$                     | Popp, 1992a   |
| <b>Prokaryotes</b>              |                                   |   |
| Symbiotic bacteria              | <i>Photobacterium phosphoreum</i> | Kobayashi et al., 1998                                    |
| Nitrogen-fixating symbiont      | <i>Bradyrhizobium japonicum</i>   | Shen et al., 1993   |
| <b>Eukaryotes, unicellular</b>  |                                   |   |
| Umbrella or cap algae           | <i>Acetabularia acetabulum</i>    | Popp, 1992a   |
| Dinoflagellate                  | <i>Prorocentrum elegans</i>       | Popp, 1992a   |
| Dinoflagellate                  | <i>Gonyaulax polyedra</i>         | Popp, 1992a; Gu, 1995; Chang, 2008a                       |
| Slime mold (also multicellular) | <i>Dictyostelium discoideum</i>   | Kobayashi and Inaba, 2000                                 |
| Algae-mushroom symbiont         |                                   |   |
| Lichen                          | <i>Parmelia physodes</i>          | Schirmacher, 2008   |
| Lichen                          | <i>Parmelia tinctorum</i>         | Bajpai, 2004, 2005a                                       |
| Lichen                          | <i>Parmelinella wallichiana</i>   | Bajpai, 2005b, 2007                                       |
| Lichen                          | <i>Xanthoria parietina</i>        | Bajpai, 2008  |
| <b>Plants</b>                   |                                   |   |
| Silver fir                      | <i>Abies alba</i>                 | Schirmacher, 2008   |
| Arabica coffee                  | <i>Coffea arabica</i>             | Gallep et al., 2004                                       |
| Robusta coffee                  | <i>Coffea canephora</i>           | Gallep et al., 2004                                       |
| cucumber seedlings              | <i>Cucumis sativus</i>            | Popp et al., 1981; Shen et al., 1993                      |
| Cucumber                        | <i>Cucumis sativus</i>            | Gu, 1995  |
| Elder bush leaflet              | <i>Sambucus sp.</i>               | Popp and Shen, 1998                                       |
| Banyan tree                     | <i>Ficus microcarpa</i>           | Schirmacher, 2008   |
| gum tree (rubber plant)         | <i>Ficus elastica</i>             | Gu, 1998  |
| mungbean seedlings              | <i>Phaseolus aureus</i>           | Shen et al., 1993; Popp et al., 1994; Popp and Shen, 1998 |

**Table 1.** List of experimental works studying photocount statistics of biological UPE related sources from different species from bacteria to human.

|                                      |                                  |  |
|--------------------------------------|----------------------------------|--|
| Purple leaf plum                     | <i>Prunus cerasifera</i> ‘Nigra’ | Schirmacher, 2008                          |
| Oak                                  | <i>Quercus robur</i>             | Schirmacher, 2008                          |
| soybean seedlings                    | <i>Glycine max</i>               | Popp, 1992a; Popp et al., 1994             |
| Soybeans                             | <i>Glycine max</i>               | Chang and Popp, 1998                       |
| Stinging nettle                      | <i>Urtica dioica</i>             | Schirmacher, 2008                          |
| <b>Animals</b>                       |                                  |  |
| Waterfleas (Crustacean)              | <i>Daphnia sp.</i>               | Popp, 1992b; Gu, 1995; Gallep et al., 2007 |
| Fireflies (Insects)                  | <i>Lampyridae</i>                | Chang, 2008a                               |
| Thailand firefly (Insects)           | <i>Lampyridae</i>                | Popp, 1992a                                |
| Chicken embryo, brain                | <i>Gallus gallus domesticus</i>  | Chang, 2008a                               |
| <b>Human</b>                         |                                  |  |
| Body                                 | <i>Homo sapiens sapiens</i>      | van Wijk et al., 2006b                     |
| Body of meditating subjects          | <i>Homo sapiens sapiens</i>      | van Wijk et al., 2008                      |
| Hand of a multiple sclerosis patient | <i>Homo sapiens sapiens</i>      | Bajpai and Drexel, 2008                    |
| Hands                                | <i>Homo sapiens sapiens</i>      | van Wijk et al., 2010                      |

Table 1. Continued

4.1. Non-biological sources

Photocount statistics measurement of weak luminescent sources was performed for solid-state ZnS:Cu luminophores (Konak et al., 1982), luminescent glass (Konak et al., 1982), and single molecules in microdroplets (Hill et al., 1998). All these experiments were analyzed in terms of thermal or Poisson statistics. The photocount statistics of diodes was found to be either Poissonian (Kobayashi et al., 1998) (for LED) or super-Poissonian (Huang et al., 2005) (for avalanche photodiodes). The chemiluminescence of a standard chemical reaction shows Poisson statistics (Collinson and Wightman, 1995). These findings prove that also random light (there is no reason to expect coherent light from *e.g.* chemical reactions or glass luminescence) can manifest photocount distribution close to Poissonian, as predicted by the theory of thermal states.

For the following discussion, it is important to stress again that Poisson statistics is not a proof of the existence of a coherent state of light. For example the superposition of a large number of independent equilibrium renewal

processes, each with a small intensity, behaves asymptotically like a Poisson process (Lindner, 2006).

## 4.2. Biological sources

There are not many works on the photocount statistics of UPE that are at the level of the quantum optics literature, without over-interpretation of the results. As an example, Kobayashi et al. (1998) is a careful and useful investigation of the photocount statistics of a time-dependent system. The authors measured the photoluminescent bacterium *Photobacterium phosphoreum* and observed a Fano factor significantly greater than one, which indicates a super-Poissonian statistics. They did not interpret this finding as an indication of a squeezed state of light but analyzed it in terms of a chaotic source (using eq. (41) of Saleh et al., 1983) with “clustering of excitation and emission”.

Another paper from the same authors represents a still more thorough investigation of the measurement of UPE photocount statistics; the experimental setup as well as possible artifacts is described in great detail (Kobayashi and Inaba, 2000). They discuss the measurement of the Fano factor in the presence of dark current and for a time-dependent source. They measured the photon statistics of *Dictyostelium discoideum* and observed the variation of the Fano factor during the early stage of development and after starvation. They found super-Poisson statistics (*i.e.* photocount distribution with a width greater than a Poissonian distribution), which they interpreted, as in their previous work, to be caused by clustering of excitation and emission processes where the optical field is composed of a sequence of independent flashes initiated by Poisson random time events. No relation to squeezed states, which can also manifest super-Poisson statistics, was mentioned. This article represents a quality benchmark for all UPE photocount measurements in terms of careful verification of the experimental setup and rigorous interpretation of the data. Kobayashi and coll. also discuss the measurement of photocount statistics with CCD cameras (Kobayashi et al., 1999; Kobayashi, 2003, 2005).

The third remarkable publication on this subject is the thesis by Schirmacher (2008) who focused on the detection of possible squeezed states of UPE. Half of the thesis is dedicated to (i) a theoretical analysis of the quantization of the electromagnetic field and (ii) the theory of photodetection. He also performed theoretical simulations of the influence of the number of modes and of the detection efficiency on the photocount distribution of squeezed states. He measured photon statistics in *Parmelia physodes*, *Prunus cerasifera* ‘Nigra’, *Abies alba*, *Ficus microcarpa*, *Urtica dioica*, *Quercus robur* and compared them with the light beam of a He-Ne laser. He observed only super-Poissonian statistics and did not find conclusive evidence of a quantum behavior of light.

There are several other scientists contributing to the field of UPE statistics and carefully performing and analyzing their experiments. For instance, Shen et al. measured the photocount statistics of cucumber seedlings, mungbean seedlings, rhizobium bacteroids, autooxidized luminol, laser light and randomized laser light (Shen et al., 1993). They found that the Mandel parameter of the photocount statistics of He-Ne laser, cucumber seedlings, mungbean seedlings, rhizobium bacteroids is close to 0 (compatible with a Poisson distribution). Yet, signals from photomultiplier detector noise, randomized laser beam or auto-oxidation of luminol showed a Mandel parameter higher than 0 (indicating a super-Poisson distribution). The authors openly stated that their aim was mainly to provide experimental data and discussed the issue of distinguishing the properties of light solely from photocount distributions. There are also other authors, who measured photocount statistics and fitted their results to statistical distributions coming from squeezed states of light (without assuming that the light field they measured was actually in a squeezed state). The parameters of these distributions enabled them to distinguish various samples (*i.e.* obtain “fingerprints”). For instance, van Wijk and coll. found specific UPE parameters for various parts of a human body (van Wijk et al., 2006b, 2010).

Other authors indulged themselves into more speculative interpretations. Fritz-Albert Popp pioneered the experimental work on the statistical properties of biological ultra-weak photon emission and motivated many scientists to work on this topic. However, many of his interpretations of experimental results are not consistent with the standard physical framework and, hence, are not generally accepted. Popp introduced the working hypothesis that the biological UPE originates from a biological coherent photon field that, further, is regulating biological processes (Popp and Ruth, 1977). This hypothesis was inspired by two indications. On the one hand, Popp had investigated several polycyclic hydrocarbons in order to find a correlation between their electronic properties and carcinogenic activity (Popp, 1976). He proposed that the mechanism of the action of the cancerogenic substances was the disturbance of the excitation cellular photon field at a certain energy level that is related to DNA repair (Popp 1976; Li, 1992a, p. 117). On the other hand, the general idea of coherent electrically polar vibration states in GHz–THz region in metabolically active cells, which had been postulated by Fröhlich (1968), was embraced by Popp as a theory generally supporting coherent processes in biology. He further assumed, with reference to the model of Li (1992b), that the DNA in the cells behaves as a low level excimer laser generating a coherent photon field.

From this time on, the experimental data obtained in Popp’s group have been attempted to fit the coherence theory of biological ultra-weak photon emission. In the following, we will highlight four specific points in the research work of Popp that are controversial either because they strongly de-

viate from standard biophysical concepts or because they lack generally accepted experimental evidence:

- *“DNA represents active photon stores which are governed by Bose condensation”* (Popp et al., 1981, p. 312; Popp, 1981, 1983; Popp et al., 1984; Popp, 1986b,a, 1995). Whereas DNA is known to be an auto-fluorescent substance, direct storage of photons on longer time scales (minutes, days) is not substantiated. Rattemeyer et al. (1981) performed experiments where DNA manifested different delayed luminescent intensities with different concentrations of ethidium bromide intercalator causing the unwinding of DNA and used these results as a proof of DNA photon storage. However, ethidium bromide is itself a fluorescent substance (Zhang et al., 2012) so it is hard to draw any conclusions from this experiment. Besides, no other group ever reproduced that result.

- *“The Poisson photocount distribution of photons detected from biological systems is a signature of a coherent field.”* Popp was well aware that Poisson photocount distribution can also come from a thermal field with many modes, but was claiming that an extremely strong mode-coupling is taking place in biological systems that reduces the effective number of modes  $M$  to approximately 1 (Popp, 1986b). While the general idea of mode coupling in physics and biology is not unreasonable (Swain 2006, 2008), such an extreme coupling of modes of an optical field in biological conditions is not experimentally confirmed and appears to be far-fetched.

- *“Hyperbolic decay of delayed luminescence is a sufficient condition for coherence”* (Li et al., 1983; Popp et al., 1984; Popp and Li, 1992; Li, 1992a; Popp and Li, 1993; Popp and Yan, 2002; Yan et al., 2005). As such, the hyperbolic decay of delayed luminescence is not generally considered to be a proof or a sufficient condition for coherence in quantum optics community. Furthermore, in some of the papers, several conceptual and mathematical mistakes were identified (see Salari and Brouder 2011, for a detailed investigation of one of these papers.) In addition, the state of light met in delayed luminescence is different from the state of light of autoluminescence (UPE) because the former is time-dependent while the latter is not. Therefore, conclusions from the study of physical parameters from measurements on delayed luminescence cannot be directly used to determine parameters of UPE.

- *“Photon emission from biological systems comes from a fully coherent electromagnetic field which serves as a basis for communication in living tissues.”* (Popp et al., 1988, p. 577). Again and as previously explained, this general claim is not substantiated by any available data.

Reviews of Popp's work can be found in some of his own papers (Popp, 2003a,b, 2005, 2009)<sup>5</sup>. He and his co-workers developed fine experimental setups, imagined clever experiments with very interesting results, but their interpretations were too far-fetched. On the one hand, this attitude took Popp away from the scientific community and brought the subject of "biophotons" into disrepute. On the other hand, F.-A. Popp's work was important in the sense that he formulated his visionary hypotheses and designed first experiments to test the field concept in biology pursuing them further to such an extent that this scientific field attracted attention of many researchers as well as the public.

The articles published by R. Bajpai focus on squeezed states instead of coherent states in order to analyze UPE photocount statistics. His main working hypothesis is that the photon field in biological systems is in a squeezed state (Bajpai et al., 1998). Squeezed states were used for modeling hyperbolic decay of delayed luminescence and photocount statistics of spontaneous UPE from lichen *Parmelia tinctorum* (Bajpai, 2004, 2005a, 2007). The squeezed state distributions provide a flexible way of analyzing UPE photocount statistics because they are mathematically described by 4 parameters with which one can fit various shapes of experimental distributions. As such, it is an interesting model. However, as for the Poisson distribution, the fact that this model fits experimental data does not necessarily mean that UPE is in a squeezed state. It is generally impossible to deduce the state of light from a photocount distribution. Higher order correlation functions of light must be measured as well, using e.g. Hanbury-Brown-Twiss-like interferometer, in order to provide evidence for squeezed or other non-classical states of light. Yet, as such, UPE statistics is an ongoing research branch and can definitely bring interesting results, when carefully elaborated.

With a background in quantum optics, Gu describes (1995) the source of UPE as a three-energy level system. He introduces super-radiance and a model involving the sum of two coherent states (Gu, 1995). Gu, furthermore, discusses non-classical light and wonders whether there are ... *nonclassical effects in biological systems*. He, furthermore, considers the biophoton field as having the property ... *to ensure an extremely high efficiency of informational transfer in life activity* (Gu, 1998). These are stimulating statements but again just speculations.

Chang (2008a,b) describes several coincidence-counting experiments (i.e. experiments with two detectors setups which aim to detect simultaneous photon emission from a single source). Photocount distributions were measured for *Dinoflagellates*, chicken embryos and fireflies *Lampyridae*.

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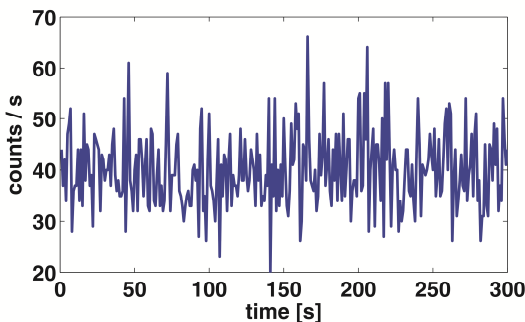
<sup>5</sup> Fritz-Albert Popp is not active in research anymore and his laboratory, International Institute of Biophysics, near Neuss, Germany, has been closed.

While the results as such are interesting and suggest all types of Poisson-distributions (namely normal, super- and even sub-Poisson photon statistics), her interpretations also contain speculative and unsubstantiated statements.

## 5. Fractal and chaotic properties of UPE time series

Independently from its physical photonic nature, UPE can also be viewed as a time series signal. Therefore signal analysis methods for time series that are used in other research fields, can be applied to UPE signals as well.

The signal parameters of UPE originate from the physical properties of light and the biochemical dynamics, both being intrinsically connected at the molecular and atomic level. From a biochemical point of view, biological processes, which generate UPE are chemical reactions involving reactive oxygen species and free radicals (see Chapter 6 of this book). Chemical and biochemical reactions can exhibit time and space periodic oscillations (Epstein and Showalter, 1996; Epstein et al., 1983; Savi, 2005; Lloyd, 2005, 2009), they can be pulsating as well as displaying complex chaotic and/or fractal dynamics (Kopelman et al., 1988; Aon et al., 2000; Benichou et al., 2010; Kopelman, 2010). Thus, it is natural to expect that also biological UPE could exhibit oscillatory and chaotic fractal behavior. Indeed, it has been demonstrated that chemiluminescence due to a Maillard-aminocarbonyl-reaction in aqueous solutions undergoes periodic and aperiodic oscillations in time (Voeikov et al., 2001 a,b).



**Figure 4.** UPE time series signals are taken from a 7 cm<sup>2</sup> surface of germinating mung beans. The signal has an appearance of a random signal. The average dark count of the detector was 13 counts/s (detector H7360-01 PMT module).

Beloussov has shown that the photon emission signals from developing loach embryos exhibit specific frequency and autocorrelation patterns (Bel-

oussov et al., 2003, 2002a,b). Such clear periodic dynamics in UPE are extremely interesting. Contrary to the usual meaning of the word, chaos is in mathematical terms a ... *Complex output that mimics random behaviour that is generated by a simple, deterministic system* (Liebovitch, 1998, p. 124). Chaotic and fractal dynamics are found in chemical and biochemical systems but manifest themselves also on the level of organisms (Stanley et al., 1999; Ivanov et al., 1999). Obviously, biological systems exhibit higher structural complexity than simple homogenous chemical systems. Biological structural complexity described in terms of fractals can be found in the literature from the level of proteins (Tejera et al., 2009) and organelles (Keough et al., 2011) to the level of physiological systems (Mainster, 1990; Liebovitch, 1998). As it is very natural to assume that the processes occurring within fractal geometrical landscapes will also manifest fractal dynamics<sup>6</sup>, it became a straightforward concept for several authors to apply the principles of fractal theory to biological UPE signals. The Fano factor  $F(T)$  (where we explicitly denote the duration  $T$  of the measurement window) is one of the simple measures to assess whether the signal manifests fractality (Teich, 1989). For a random Poisson process in which fluctuations in photon counts are uncorrelated,  $F(T)$  is approximately 1 for all window sizes (Teich, 1989, 1992). For a fractal process,  $F(T)$  increases as a power of the window size and may reach values greater than 1 (Teich, 1989, 1992). The slope of the doubly logarithmic plot of  $F(T)$  is the scaling exponent, often denoted as  $\alpha$ , which is the power to which fluctuations in photon counts on one time scale relate to those on longer time scales. The scaling exponent is useful for assessing self-similarity, one of the features of fractal signals.

The Fano factor as well as the first four statistical moments (mean, variance, skewness, kurtosis) of the photocount distribution have been used by the group of Van Wijk to characterize the UPE signal from three body locations of a single human subject (van Wijk et al., 2006a); they found that the Fano factor as well as statistical moments were different for each body location. Studies in this direction were further developed because authors considered the Fano factor to be a useful parameter to fingerprint the UPE signal. UPE signals from the dorsal and palmar side of both hands of 50 human subjects (van Wijk et al., 2010) and from two pre- and post-meditating human subjects (Van Wijk et al., 2005) have been measured and analyzed for  $F(T)$ , statistical moments and doubly logarithmic plot of  $F(T)$ . Fifty human subjects showed  $F(T) = 1$  for  $T < 3$  s and rather large variation of their

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<sup>6</sup> Good example of relation of structural and dynamical fractality can be seen in the heart (West and Deering, 1994), where the His-Purkyne system which innervates the myocard has geometrically fractal branching. Some authors (West and Deering, 1994) stipulated that the fractal branching of the electrical depolarization wave results in fractal scaling in dynamics of heart rate variability.

$F(T)$  for  $T$  greater than a few seconds. A difference in  $F(T)$  of UPE from pre- and post-meditating human subjects has been observed. Furthermore, a doubly logarithmic plot of  $F(T)$  dependence has also been used (van Wijk et al., 2011) to characterize UPE signals of respiratory bursting neutrophils. The authors suggested that Fano factor analysis could provide information regarding leukocyte interactions because any deviation from Poisson statistics contains information within the sequence of photon counts events about the cell population as a collective phenomenon.

However, using Fano factor without signal preprocessing and careful observation of the possible technogenic origin of UPE signal fluctuation can be tricky. Fano factor of the background (detector) noise, which can be generally different from that of the UPE signals, needs to be taken into account especially for those cases where the signal to noise ratio of UPE is low. For example, one can observe different  $F(T)$  of two statistically identical UPE signals simply because one has a lower intensity and is closer to the noise level of the detector. The Fano-factor itself is only a good measure for signals without any decreasing or increasing trend, for otherwise it will yield misleading results. Since shuffling (randomization) of data with decreasing or increasing trend removes both the trend and long-range correlations, it cannot be generally used as a surrogate signal. There exist a number of other and much more developed methods to free a signal from trends and obtain a more reliable quantification of fractal and chaotic parameters of the signal (Stanley et al., 1999). Scholkmann et al. (2011) pioneered the use of more advanced fractal analysis methods on UPE signals: Multifractal detrended moving average analysis of UPE signals from germinating wheat seedlings was used to differentiate between two grades of intoxication with potassium dichromate.

It is essential to extend the advanced signal analysis of UPE signals to see if the obtained parameters can have a differentiating and diagnostic “fingerprint” character.

## 6. Conclusion

The conclusion of our review is that, up to now, no reliable estimate of the coherence of UPE was made by the different methods of photocount statistics measurement. The presence of coherence seems to follow from a straightforward reasoning: a living organism must be in some coherent state because it is obviously not in thermal equilibrium (del Giudice et al., 2005). However, the actual situation is more subtle: on the one hand, a thermal source can emit partially coherent light, even close to the source (Greffet et al., 2002), and, furthermore, independent thermal sources can

produce two-photon interference (Zhai et al., 2006)<sup>7</sup> while, on the other hand, the organization required to maintain life has no *a priori* reason to imply that UPE is in a coherent state. Moreover, thermal states and coherent states are two extremes of a very broad range of possible states of light. What we would need is to actually measure the coherence length and time of UPE. The UPE coherence times given by Popp (10 days<sup>8</sup>) and Bajpai (5 hours<sup>9</sup>) seem to be completely off the mark. Standard methods in quantum optics can deliver more reliable information on coherence and statistical properties of UPE of living systems. Coherence parameters could be quantified by measuring light interferences or light correlation functions. A non-classical, *i.e.* quantum nature could be assessed by using a Hanbury-Brown-Twiss interferometer, but the extremely low intensity of UPE makes these experiments highly challenging. However, for the concept of coherence in biology as such, it needs to be noted that the coherence of vibrational and spin dynamics in biomolecules is gaining acceptance among biophysicists (Cimei et al., 2002; Gruia et al., 2008; Liebl et al., 1999; Engel et al., 2007; Parson, 2007; Wolynes, 2009).

Furthermore, there are indications that other signal properties of biological UPE than those studied in quantum optics, such as coherence, are naturally oscillatory, complex (chaotic) and fractal. Thus, suitable methods adapted from statistical physics and already used for other biological signals to uncover “hidden information” (Goldberger et al., 2002) may be also used to analyze the UPE signals.

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<sup>7</sup> One should keep in mind that two-photon interference is not the interference of two photons (Pittman et al., 1996).

<sup>8</sup> “A reasonable coherence time is the lifetime of cell organelles (for instance, mitotic figures) of about ten days” (Popp, 2009, p. 59).

<sup>9</sup> “The signal was, therefore, coherent for 5 hr” (Bajpai, 1999).

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