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Coherent Nature of the Radiation Emitted in Delayed Luminescence of Leaves

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After exposure to light, a living system emits a photon signal of characteristic shape. The signal has a small decay region and a long tail region. The flux of photons in the decay region changes by 2 to 3 orders of magnitude, but remains almost constant in the tail region. The decaying part is attributed to delayed luminescence and the constant part to ultra-weak luminescence. Biophoton emission is the common name given to both kinds of luminescence, and photons emitted are called biophotons. The decay character of the biophoton signal is not exponential, which is suggestive of a coherent signal. We sought to establish the coherent nature by measuring the conditional probability of zero photon detection in a small interval Δ. Our measurements establish the coherent nature of biophotons emitted by different leaves at various temperatures in the range 15–50°C. Our set up could measure the conditional probability for $\Delta \leqslant 100$ μs in only 100 ms, which enabled us to make its measurement in the decaying part of the signal. Various measurements were repeated 2000 times in contiguous intervals, which determined the dependence of the conditional probability on signal strength. The observed conditional probabilities at different signal strengths are in agreement with the predictions for coherent photons. The agreement is impressive at the discriminatory range, 0.1–5 counts per Δ, of signal strengths. The predictions for coherent and thermal photons differ substantially in this range. We used the values of Δ in the range, 10 μs–10 ms for obtaining a discriminatory signal strength in different regions of a decaying signal. These measurements establish the coherent nature of photons in all regions of a biophoton signal from 10 ms to 5 hr. We have checked the efficacy of our method by measuring the conditional probability of zero-photon detection in the radiation of a light emitting diode along with a leaf for Δ in the range 10 μs–100 μs. The conditional probability in the diode radiation was different from that predicted for coherent photons when signal strength was less than 2.5 counts per Δ. Only the diode radiation exhibited photon bunching at signal strength of around 0.05 count in Δ.

1. Introduction

Delayed luminescence is a phenomenon of photon emission by a living system after its stimulation by visible radiation. Strehler & Arnold (1951) first observed it as an afterglow in the green plants exposed to light illumination. Lavoral (1975) identified three separate phases in the afterglow and characterized them by the relaxation time τ. The phases are fast ($\tau < 1$ μs), medium (1 μs $< \tau <$ 1 ms) and slow ($\tau > 1$ ms). The signals of the fast and medium phases are

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intense but they decay rapidly and become negligible in a few milliseconds. The signal of the slow phase is very weak but persists a little longer. It is called delayed luminescence (Jursinic, 1986). The phenomenon of delayed luminescence is not restricted to green plants or photosynthetic systems alone but has been observed in many biological systems from bacteria to human tissues (Popp *et al*., 1988). A biological system after a few seconds exposure to normal laboratory illumination, starts emitting the full strength delayed luminescence signal. The signal is observable for duration ranging from tens of milliseconds to a few minutes in different systems. The origin and purpose of the phenomenon are not known. Living systems also exhibit another kind of luminescence with intensity much weaker than the peak intensity of delayed luminescence. It is called ultra-weak luminescence. It is equally wide spread (Ruth, 1989) and is observable for a much longer time. Its origin and purpose are also not known. The photons emitted in both kinds of luminescence are mainly in the visible range and are detected by a broad band scintillation counter sensitive in the range 300–800 nm. Both kinds of luminescence go under the common name of biophoton emission and the associated photons are called biophotons (Popp, 1992). The new name emphasizes their biological connection and implies the same or similar nature. The two kinds of luminescence differ in one respect—the signal of delayed luminescence decays and becomes unobservable in a small time after stimulation while the signal of ultra-weak luminescence is non-decaying and is observable throughout the lifetime of a system. Delayed luminescence is considered light-induced and is called light-induced biophoton emission. Ultra-weak luminescence is supposed to be unconnected with light stimulation and is called spontaneous biophoton emission (Popp, 1992). The spontaneous nature of ultra-weak luminescence is difficult to establish because of the weakness and long duration of the signal. One can not rule out the emission of a long-tailed light-induced biophoton signal with a non-decaying part that is observed as ultra-weak luminescence (Bajpai & Bajpai, 1992).

The broad features of the shape identify a biophoton signal, while its specific features characterize the emitting system. The shape exhibits a sensitive dependence on many environmental and physiological factors (Popp *et al*., 1988). The dependence on different factors has not been quantified so far and it has not been possible to disentangle their contributions in a consistent manner in the conventional framework of photon emission. The difficulties in understanding the shape force us to suspect that the origin of biophotons lies beyond the conventional framework. The determination of the nature of biophotons may confirm the suspicion. We, therefore, planned to determine the nature of photons in a biophoton signal by measuring the conditional probability of zero-photon detection in a small interval after the detection of a photon. We have measured the conditional probability and its dependence on signal strength. Our measurements establish the coherent nature of biophotons and rules out their description in the conventional framework.

The inadequacy of the conventional framework to describe a biophoton signal is easy to demonstrate in the semi-classical picture. The semi-classical picture describes photons by their classical radiation field and the emitting system by the quantum states of its sub-units (Weisskopf & Wigner, 1930). A sub-unit can be an atom, a molecule, a bio-molecule, or any other substructure. The sub-unit emits a photon in its de-excitation from a higher energy state to a lower energy state. Each de-excitation is a spontaneous quantum jump of probabilistic nature. One can define the transition probability per unit time of a de-excitation, that is specific to the sub-unit and can be taken to be a constant. The transition probability gives the lifetime of the excited state (Fonda *et al*., 1978). A photon signal contains a large number of photons which are emitted in the different de-excitations of various sub-units. The conventional framework envisages all de-excitations to be independent. The separate identity of sub-units and the independence of de-excitations give rise to the thermal nature of photons and exponential decay character of signals. Photons of thermal nature are called thermal photons. Phase, amplitude, and intensity of the classical radiation field of thermal photons fluctuate in time in an

incoherent manner and the statistics of counts detected in a fixed time interval in the signal of constant strength is given by a thermal distribution (Loudon, 1975). The incoherent nature of photons emanates from the independence of de-excitations, and their thermal distribution over a large number of de-excitations. The exponential decay character of the signal is merely a consequence of the probabilistic nature of independent quantum jumps. Quantum jumps between the same pair of states give rise to a mode of the signal. Each mode is of definite wavelength and decays exponentially. A signal may have many modes but all modes are uncoupled because of the independence of various de-excitations. The conventional framework does not allow a non-decaying mode or a mode with a non-exponential decay character. It also does not envisage any relationship among decay constants of different modes. Hence, it can not describe a spontaneous biophoton signal that does not decay at all. Neither can it describe a light-induced biophoton signal, whose exponential decomposition does not yield satisfactory lifetimes. Popp (1992) has analysed many light-induced biophoton signals and noticed that the exponential decomposition of a signal requires many modes whose number varies with environmental and physiological conditions; and the decay constants are situation specific and unrelated to the known lifetimes. The exponential decomposition was devoid of any physical content. Popp & Li (1993) also found that a single hyperbolic decay term gave a better representation of the biophoton signal.

The conventional framework with a few additional assumptions does allow the emission of signals of desired shapes. A non-decaying signal requires an additional source of energy that replenishes the energy radiated by a sub-unit, while a hyperbolic decaying signal requires either a singular transition probability or a specific relation among the decay constants of many modes. These assumptions retain the semi-classical picture; the sub-units de-excite independently and the nature of photons remains thermal. Our observation of the coherent nature of biophotons shows the futility of these additional assumptions and the inadequacy of the conventional framework. The emission of a signal of coherent nature requires a cooperative de-excitation of various sub-units. The cooperative de-excitation obliterates the separate identity of sub-units and is against the basic philosophy of the conventional framework. It invokes a new framework in which the living system emits photons as a single unit. Photons emitted by a single unit will not be thermally distributed and will be in a pure quantum state. Photons emitted in a pure quantum state have a coherent nature and are generically called coherent photons (Loudon, 1975). Coherent photons exhibit non-classical effects. Phase, amplitude, and intensity of the classical radiation field associated with coherent photons do not fluctuate. Coherent photons, therefore, exhibit interference effects as well. The distribution of photo-counts in a signal of coherent photons depends upon the quantum state of the photons and is not thermal. We expect biophotons to possess all the properties of coherent photons. The exceptional shape of a biophoton signal is also not problematic in the new framework. It arises naturally from the dynamic evolution of the quantum state of coherent photons. One needs a model to specify the state and its evolution. We have demonstrated the method of determining the shape in a prototype model (Bajpai *et al*., 1998). The model assumes the quantum state of biophotons to be a squeezed state (Yuen, 1976) whose dynamics is given by a frequency stable damped harmonic oscillator. The shape of a biophoton signal in the model is given by a sum of three terms with constant, hyperbolic decay and inverse square decay behaviours. The strength of each term is situation and system specific. The model can explain the shape of a signal, its specificity and its dependence on physiological and environmental factors.

The coherent nature of biophotons is desirable in another context. All holistic models (Beloussov & Popp, 1995) postulate the existence of a wave field in and around a living system. The field is used for the transfer of information and instructions and is required to be coherent to ensure the cooperative functioning of spatially separated parts of a living system. Such a holistic field has not been identified so far. The ubiquitous presence of biophotons does suggest a possibility of identifying them as the quanta of

a holistic field. Their coherent nature will then be a manifestation of the coherence of holistic field. Li (1992) has even speculated that the quantum state of biophotons has to be a coherent or squeezed state, because these states can transfer information in an almost loss-less manner. A quantum squeezed state can arise from another speculation that bestows on the living system the capability to detect an object (or objects) whose interaction is weaker than the thermal noise. Such a capability will confer an evolutionary advantage to the living system. The system acquires the capability by performing continuous quantum non-demolition measurements on some photon field (Bocko & Onofrio, 1996). The system may start with any initial field, but the first quantum non-demolition measurement will collapse the wave function of the initial field into a squeezed state and the wave function will remain in the squeezed state after subsequent measurements. These are interesting speculations, which have motivated various attempts to detect the coherence of biophoton signals.

Experiments performed in the past to detect the coherence of biophoton signals were of three different types. In the experiments of the first type, the intensity of a biophoton signal was measured at different cell concentrations. The intensity of the signal did not vary linearly with cell concentration, but showed an oscillatory character (Galle *et al*., 1991). The oscillatory character is a typical interference effect produced only by coherent waves. The experiments, therefore, showed that different cells were emitting biophotons of coherent nature. In the experiments of the second type, the attenuation in the intensity of signals passing through a dispersive medium was measured. The attenuation in a biophoton signal was at least an order of magnitude smaller than the attenuation in an incoherent photon signal of similar intensity (Popp & Deny, 1991). The attenuation in the intensity is caused by scattering losses, which are reduced in a coherent signal having a precisely defined intensity and direction of propagation. The smaller attenuation in the biophoton signal found in the experiments was an indication of the coherent nature of biophotons. In the third type of experiment, the statistical distribution of photo-counts was measured. The statistical distribution of photo-counts was nearly Poissonnian in the majority of biophoton signals; there were also a few cases of sub- and super-Poissonnian distributions (Popp *et al*., 1988). The statistical distribution of photons detected in a fixed time interval depends upon the state of photons. The distribution is Poissonnian if photons are in a quantum coherent state; sub-, super- or simply Poissonnian if photons are in a squeezed state (Yuen, 1976); and a Planck distribution if photons are in a thermal equilibrium state (Perina, 1985). The observed distributions suggested that biophotons were in a pure quantum state. In a few cases, the quantum state was probably a squeezed state showing sub- and super-Poisson distributions. The observed photo-counts are not always able to discriminate among various statistical distributions. The identification of the correct statistical distribution is very difficult in strong signals, while the measurements are erroneous in weak signals because of the background noise. Only the signals with strength of around 1 count in the measuring interval can effectively discriminate between thermal and Poisson distributions. The noise in the experimental setup was nearly 1 count per 100 ms and the intervals used in the measurements were longer than 100 ms. The discriminating signals, therefore, contained a substantial amount of the background noise. The noise introduced ambiguities in the determination of statistical distribution and could provide suggestive evidence of the coherent nature. One has to perform some other type of experiment to obtain conclusive evidence of the coherent nature.

The correlation among photons detected at different times in a signal depends upon the nature of photons and its measurement in biophoton signals can provide direct and conclusive evidence of their coherent and quantum nature (Perina, 1985). The correlation is measured by various conditional probabilities. The conditional probability of zero photon detection (or equivalently of at least one photon detection) in a time interval Δ after the detection of a photon is an easily measurable conditional probability that can discriminate between thermal and coherent photons. It is very effective if the signal strength is around 1 count in Δ. It also

exhibits photon bunching—its value for thermal photons becoming twice the value for coherent photons when signal strength goes to zero. Arecchi *et al*. (1966) used its measurements to differentiate between coherent photons of laser radiation and quasi-thermal photons. We also planned to measure it in biophoton signals using an improved technique that reduced background noise and other experimental errors. Our technique has two improvements—use of a fast electronic data acquisition system and continuous repetitions of a measurement. The electronic data acquisition system allowed us to reduce the duration of a single event to 1 μs in a normal measurement and to 10 μs in a coincidence measurement. The reduction in the duration of a single event reduced the background noise to a negligible level in discriminating signals and allowed us to determine the photo-count statistics and conditional probability only in 100 ms. One could ignore in this duration the decay of a delayed luminescence signal with characteristic decay time $\tau > 10$ s. This enabled us to determine the photo-count statistics and conditional probability of zero-photon detection in the decay region of a signal. Since a signal is more intense in the decay region, the contribution of the background noise is much smaller. The electronic data acquisition system could repeat a measurement in any number of contiguous intervals. The repeated measurements in a decaying signal determined the dependence of the conditional probability on signal strength. The dependence was in the form of a smooth curve that was almost free from the errors of individual measurements.

2. Materials and Methods

We used the experimental set up (Popp *et al*., 1994) fabricated by Professor Popp for investigating the role of biophotons in signal communication between living systems (Ho *et al*., 1994). The details of the experimental set up and measuring procedure are given in Popp *et al*. (1994). The measuring system consisted of two photo-multiplier tubes (EMI 9558 QA) placed 12.5 cm apart, two counters and a coincidence device. There were two separate chambers for two different samples. One could detect photons emitted by a sample source in a photo-multiplier tube and register them in a counter. The entire setup consisting of a source, its detector and counter were called a channel. There were two identical channels, which independently registered the photons emitted by two different sources. A coincidence device (DESY Lab DAP-DLL) coupled the channels asymmetrically. One channel was identified as photon channel and the other as reference channel. (Either of the two channels could be identified as photon channel.) The coincidence device also had a counter that registered the counts of the reference channel but only for a time interval Δ after each count in the photon channel. Let N, R, and Z be the number of counts registered, respectively, in the photon channel, reference channel, and coincidence device during a time T. These numbers directly measure signal strength $n(\Delta)$ and the conditional probability $P_0(\Delta)$ of zero photon detection in an interval Δ in the photon channel. The signal strength is given by

$$n(\Delta) = N\frac{\Delta}{T}, \tag{1}$$

and the conditional probability by

$$P_0(\Delta) = \left(1 - \frac{Z}{R}\right). \tag{2}$$

The expression for $P_0(\Delta)$ requires an explanation. The right-hand side of eqn (2) gives the fraction of counts of the reference channel not registered in the coincidence device. If the value of R is large, say more than 100, then $P_0(\Delta)$ is equal to the probability of not registering a count of the reference channel in the coincidence device. The coincidence device does not register a count only if no photon was detected in the photon channel in the prior time interval Δ. Consequently, the probability of not registering a count in the coincidence device is equal to the conditional probability of not detecting a photon in the prior time interval Δ. The conditional probability of not detecting a photon in the prior time interval Δ is equal to the conditional probability of not detecting a photon in the subsequent time interval Δ because of time reversal invariance. This is the result given in eqn (2). The experimental setup is able to measure in a time

T the cumulative outcome of N events. Although the duration of each event is Δ, T is, in general, not equal to $N\,\Delta$. This is because one does not have to wait for the entire duration of an event to know its outcome. The detection of a photon within the interval Δ gives the outcome of the ongoing event. The detection also initiates a new event. If it is further assumed that the photon emission is a Poisson process and the number of counts of the photon channel is proportional to the number of photons emitted by the source, then $P_0(\Delta)$ is also equal to the probability of not emitting a photon in an interval Δ. The probability of photon emission in the same interval Δ will given by $(1 - P_0(\Delta))$, and is equal to Z/R—the fraction of counts actually registered in the device. It is pointed out that there is no need of a detector and a photon source in the reference channel. One could replace them by a signal generator. We have used both of them in our measurements.

R, T, and Δ were the adjustable parameters of a measurement. R was adjusted by changing the strength of the source in the reference channel (or by changing the frequency of the signal generator). A large value of R reduces the fluctuations in the measured ratio Z/R; a value of R greater than 100 gave satisfactory results. The parameters T and Δ were fixed by hand at the start of a measurement. The values of the parameters could be varied in steps of 10 μs. We imposed two inequalities on the choice of these parameters: $\Delta \ll T$ and $T \ll \tau$. The first inequality ensured the inclusion of a large number of events in a determination of the probability, while the second inequality ensured their similarity. We usually took $T \geqslant 1000\Delta$ and thus determined the probability from the outcomes of more than 1000 similar events. We used $T = 100$ ms in the decay region of a biophoton signal where $\tau \sim 10$ s, and $T \leqslant 10$ s in the tail region where the signal was almost constant. The choice of the parameter Δ is more crucial for it determines the signal strength. Its value was chosen so as to obtain the discriminating range of signal strength in any desired region of a signal. The values of Δ in the range 10 μs–10 ms gave the desired signal strength in the various regions of a signal from 10 ms to more than 5 hr. The measurement of the conditional probability for each set of parameters was repeated 2000 times in contiguous intervals to obtain the dependence of the probability on signal strength.

Leaves used in the measurements were of different sizes and from different plants—both fresh ones, as well as those plucked up to one week earlier were used. A leaf was stimulated by an exposure to white mercury light for 10 s (Bajpai & Bajpai, 1992) and its biophoton signal was observed for a few minutes. The procedure was repeated to check the stability of the signal. The dependence of the probability of zero-photon emission on signal strength was measured only in the leaves emitting stable signals. The measurements were repeated at various temperatures in the range 15–50°C in a few leaves. We also measured the probability of zero-photon emission in the radiation emitted by a commercially available 3 mm As-Gl green light emitting diode, henceforth called a micro-lamp—a non-biological source of optical radiation. The strength of the signal in the micro-lamp depended linearly on the firing potential. A decaying signal was obtained by supplying the firing potential through a discharging condenser. The strength and decay constant were adjusted to obtain a signal similar in these characteristics to the biophoton signal of a leaf. The probabilities of zero-photon emission in the radiation of the lamp and of the leaf were measured one after the other under identical measuring conditions. We could not succeed in obtaining a suitable decaying signal required for the measurement of conditional probability of zero-photon detection in a purely thermal source, for which we determined only the photo-count statistics at a few discrete values of signal strengths.

3. Results

The salient features of our measurements are summarized below:

1. the values of N and R at contiguous intervals gave the shapes of the delayed luminescence signals from two different sources for 200 s or more. The shapes had the usual character—an initial hyperbolic decaying part followed by a non-decaying part. The measured

values of Z and R showed an interesting feature—the ratio Z/R depended upon N, Δ and T through a single variable $n(\Delta)$ given by eqn (1). The dependence had a universal character and was the same in all biophoton signals. We have plotted the observed values of Z/R of different leaves at various choices of measuring parameters as a function of $n(\Delta)$ in the same scatter plot. All data points are scattered around a curve that gives the dependence of the probability of photon detection in the interval Δ as a function of signal strength $n(\Delta)$. The observed curve is almost identical to the curve predicted in the Glauber theory of coherent photon detection (Glauber, 1963). The scatter plot of points obtained in a single set of continuous measurements is depicted in Fig. 1 in a limited region. The figure also depicts the predicted curves for thermal and coherent photons. The parameters of the measurements were $\Delta = 1$ ms and $T = 100$ ms; these parameters were not optimal and large fluctuations were expected. The figure indicates that most of the points are very close to the curve predicted for coherent radiation, and are quite far from the curve predicted for thermal radiation. The agreement between observed data points and the values predicted for coherent radiation was much better in cases where $T \geqslant 1000\,\Delta$;

2. the probability of photon detection varied by more than two orders of magnitude in a decaying signal. We, therefore, divided the observed probabilities by the predicted probabilities for thermal and coherent radiation, thus obtaining two probability ratios for each observation. The probability ratio for coherent radiation was mostly around 1, whilst for thermal radiation was quite often different from 1 and varied with signal strength. The probability ratios at the two ends of our measuring conditions are depicted in Figs 2 and 3. The parameters of these measurements were $\Delta = 10\,\mu$s and $T = 100$ ms and $\Delta = 10$ ms and $T = 10$ s, respectively. Figure 2 is a scatter plot, while Fig. 3 is a line plot of the data. The choice of parameters of Fig. 2 was such that the difference in the two probability ratios was perceptible only during the first 120 s. The two probability ratios became indistinguishable subsequently. The overlapping values of the two

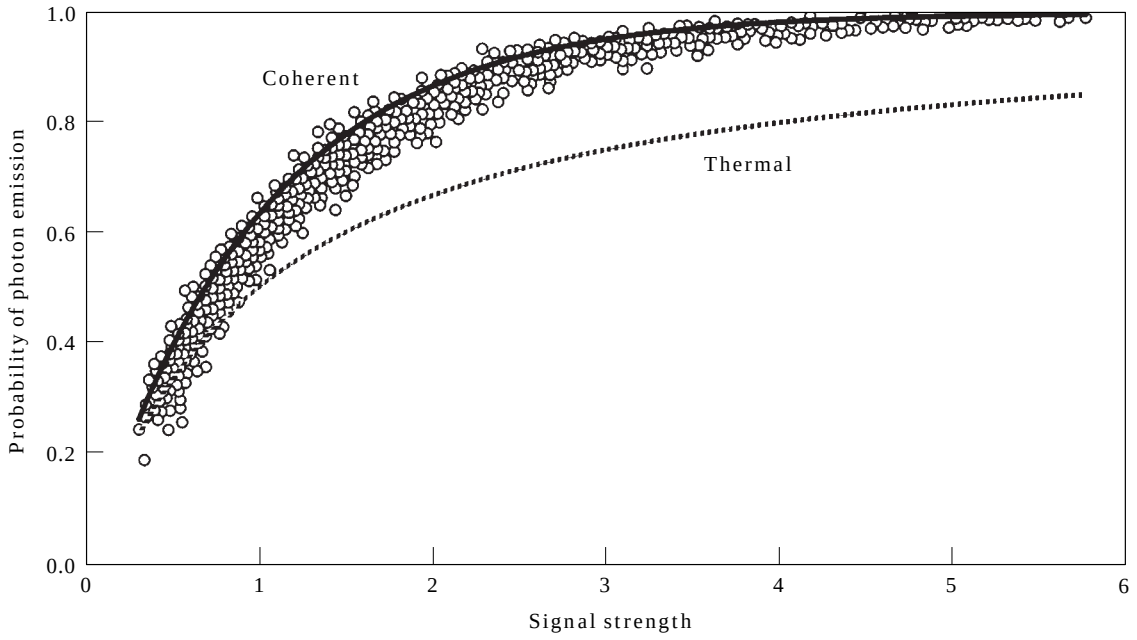

FIG. 1. Dependence of $(1 - P_0)$ on signal strength: the measured probability $(1 - P_0)$ of photon emission in 1 ms is plotted against the signal strength $n(\Delta)$. The dotted and continuous curves give, respectively, the predictions for thermal and coherent radiation. The measuring parameters were $T = 100$ ms and $\Delta = 1$ ms.

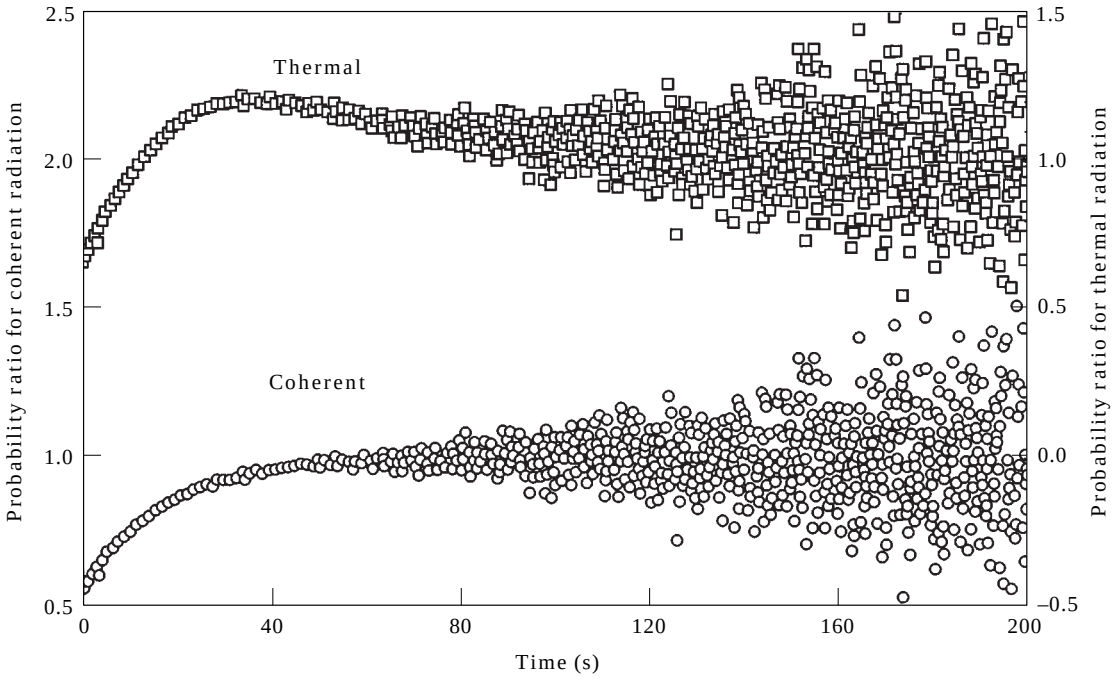

FIG. 2. Effect of non-linearity in detection: the probability ratios of photon emission in thermal and coherent radiation are plotted in a decaying signal. The right side y-axis corresponds to thermal radiation. Signal strength was more than 10^6 counts s^{-1} for 60 min. The measuring parameters were $T = 100$ ms and $\Delta = 10$ μs.

probability ratios are separated in the figure by displacing the scale of the probability ratio for thermal radiation.

Figure 2 also shows that the probability ratio for coherent radiation was slightly less than 1 during the initial 50 s of the decay. The signal was intense in this duration and showed a counting rate higher than 10^6 counts s^{-1}. We do not consider it to be indicative of incoherence and attribute it to the malfunctioning of the photo-multiplier tube and coincidence device. The detection in a photo-multiplier tube becomes nonlinear and the dead time of a coincidence device (≈ 1 μs) becomes appreciable at high counting rates. Similar behavior was observed in all intense signals. We consider the data points at counting rates higher than 10^6 counts s^{-1} to be spurious. The probability ratios of this particular leaf are depicted in the figure to highlight the apparent incoherence arising from the instrumental deficiencies. The probability ratio for coherent radiation in less intense signals was always around 1.

Figure 3 depicts the two probability ratios for $\Delta = 10$ ms and $T = 10$ s. The two probability ratios differed considerably in the tail region. We have observed them for more than 5 hr but have depicted only a part of the data. The signal strength was almost constant in the tail region and the figure depicts the fluctuations of repeated measurements. The full data set simply extends this depiction. The average probability ratio of the full data set was 1.00 ± 0.02 for coherent radiation and 1.26 ± 0.07 for thermal radiation. Figure 3 also shows a small amount of incoherence in the initial stage of the decay. The duration of the incoherence in the figure is 3 min and is much longer than 50 s shown in Fig. 2. This incoherence is also only apparent, however. We attribute it to the choice $T = 10$ s, that gives the duration of a probability measurement. It was a long duration and various events spread over this duration in the decaying part of the signal could not be considered similar. The determination of the probability was, therefore, erroneous. It gave rise to an apparent incoherence that lasted until the contribution of the decaying part became negligible.

The data of Figs 2 and 3 were obtained from the same leaf using different measuring parameters. The figures demonstrate the coherent nature of biophotons emitted at the two extremes

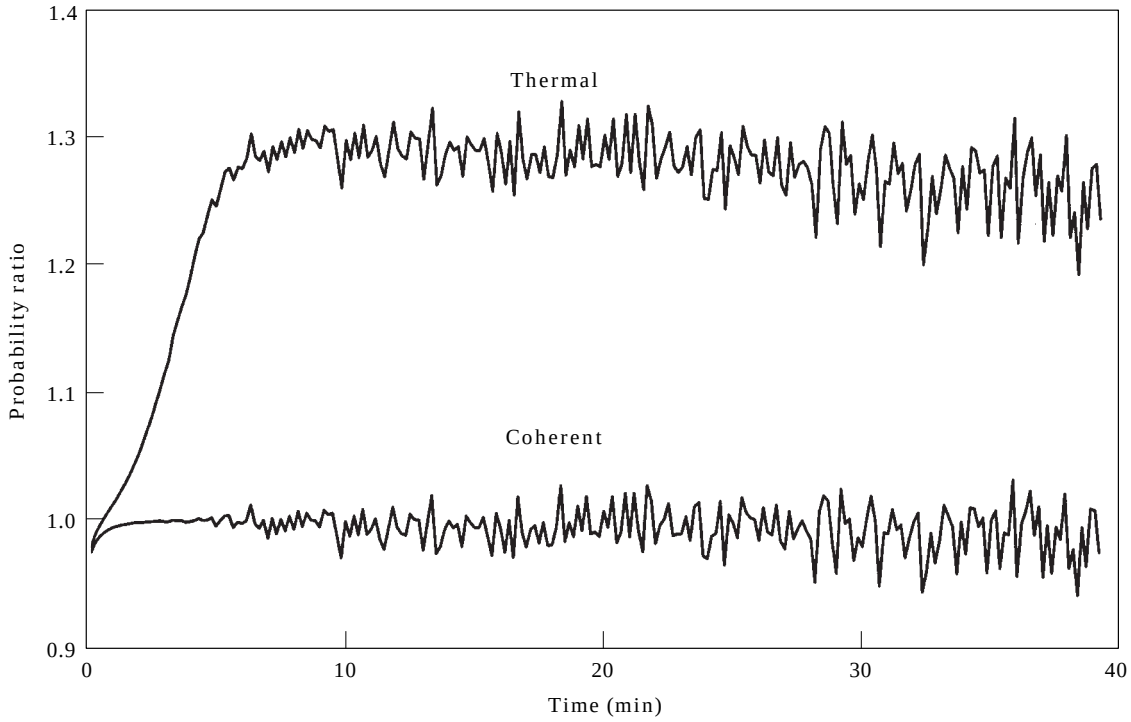

FIG. 3. Coherent nature of radiation: the ratio of observed to predicted probability ($1 - P_0$) of photon emission in thermal and coherent radiation are plotted as a function of time. The behavior is shown only up to 2250 s, being similar beyond this time. The measuring parameters were $T = 10$ s and $\Delta = 10$ ms.

of a delayed luminescence curve. The data points obtained with other choices of parameters similarly demonstrate the coherent nature in other regions of the delayed luminescence curve;

3. the overall behavior of the two probability ratios is presented in Table 1 for different choices of parameters. The table gives the average and the variance of the two ratios in different cases. The spurious data points were ignored in the calculations. The average probability ratio for coherent radiation is nearly 1 in all cases and its variance is small. In contrast, the average probability ratio for thermal radiation is not equal to 1; it varies with the choice of

TABLE 1

The probability ratios of zero-photo detection in a leaf and a micro-lamp: *the average and standard deviation of the probability ratio for coherent and thermal radiation in a leaf and for coherent radiation in a micro-lamp are given for different measuring parameters* T *and* Δ. *The number of data points in each set were* 2000, *but we have excluded from the calculations the data points with counting rate higher than* 1 *million counts* s^{-1}

S. No.	T	Δ	Leaf (coherent)	Leaf (thermal)	Lamp (coherent)
1	100 ms	10 μs	1.00 ± 0.11	1.05 ± 0.12	1.52 ± 0.26
2	100 ms	20 μs	1.00 ± 0.08	1.08 ± 0.10	1.41 ± 0.31
3	100 ms	30 μs	1.00 ± 0.06	1.11 ± 0.08	1.56 ± 0.34
4	100 ms	50 μs	0.99 ± 0.07	1.11 ± 0.10	1.27 ± 0.28
5	100 ms	100 μs	0.99 ± 0.02	1.24 ± 0.14	1.11 ± 0.21
6	1 s	1 ms	1.00 ± 0.07	1.14 ± 0.10	1.02 ± 0.17
7	10 s	10 ms	1.00 ± 0.02	1.26 ± 0.07	

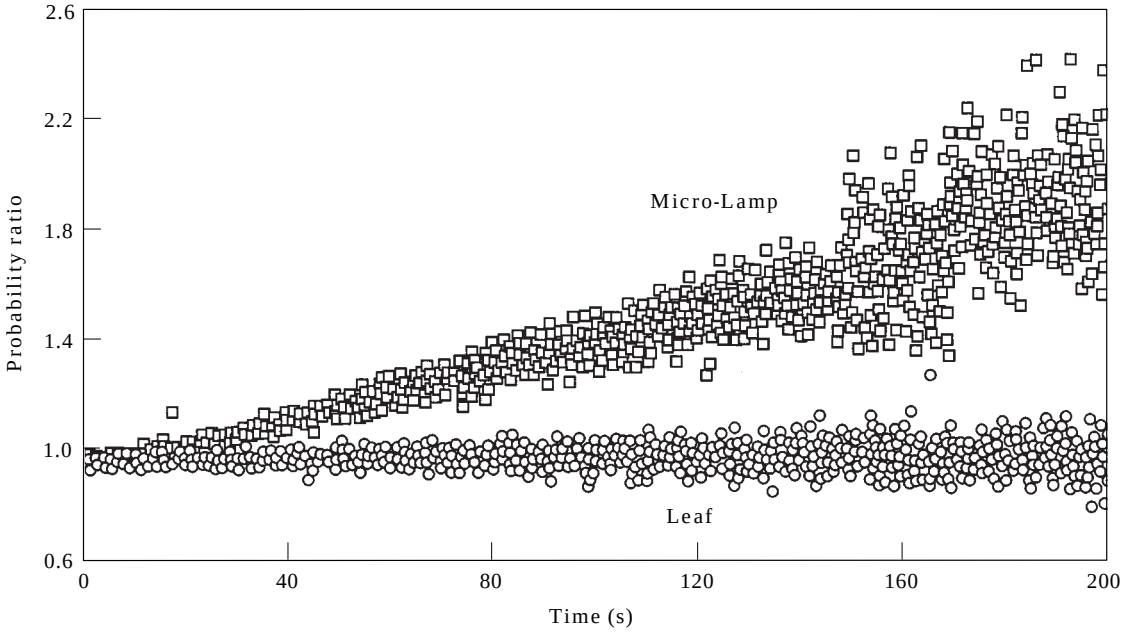

FIG. 4. Radiation from two different sources: the probability ratio of photon emission for coherent radiation from a micro-lamp and a leaf. Both sources emitted radiation of comparable strength. The measuring parameters were $T = 100$ ms and $\Delta = 20$ μs.

parameters, and depends upon the signal strength. The variance is large, particularly in cases which contained more data points in the discriminating range of signal strength. The table indicates that the probability ratio for coherent radiation was 1 at any signal strength in the biophoton signal of a leaf and that the nature of biophotons was coherent;

4. the probability ratio for coherent radiation was always around 1 in other leaves at different temperatures and in different stages of growth and decay. We measured the ratio in one leaf at different temperatures in the range 15–50°C. We also varied the time gap between a measurement and the plucking of the leaf in the range 1 min–7 days. The intensity of the signal did change with temperature and time gap, but the observed probability ratio for coherent radiation always remained nearly 1;

5. the behavior of the probability ratio for coherent radiation was compared in the signals emitted by a micro-lamp and a leaf. The probability ratio for coherent radiation of the two samples is depicted in Fig. 4. The measuring parameters were $\Delta = 20$ μs and $T = 100$ ms. The probability ratio for coherent radiation in the micro-lamp was around 1 only for about 10 s when the signal strength was more than 2.5. It then started increasing and attained the value 2 after 190 s when the signal strength became nearly 0.05. The probability ratio subsequently hovered around 2. This was a typical photon bunching effect and was not shown by the biophotons. The probability ratio of the micro-lamp also depended only upon signal strength and behaved in a predictable manner for other choices of Δ in the range $10\ \mu s \leqslant \Delta \leqslant 100\ \mu s$. The overall behavior of the probability ratio observed during 200 s for different choices of measuring parameters is also given in Table 1. The table shows that the average value of the probability ratio is significantly different from 1 for $\Delta < 100$ μs; the micro-lamp does not emit coherent photons;

6. we did not succeed in measuring the dependence of the probability of zero-photon emission on signal strength in the radiation emitted by an incandescent lamp because of the sensitivity of the signal to applied voltage. The signal strength varied approximately as the eighth power of the applied voltage. It was impossible to produce a measurable and decaying signal. We did succeed in measuring the photo-count statistics of radiation emitted by a

hot metal tip and a leaf using events of a few microseconds duration. The photo-count statistics was Poisson in biophotons and binomial in the radiation of the metal tip.

4. Discussion

The nature of biophotons is determined from the measurement of conditional probability of zero-photon detection and its dependence on signal strength. Glauber (1963) has developed the theory of single photon detection for calculating various probabilities and their dependence on signal strength. Mandel (1963) used this theory in a semi-classical framework to obtain the following expression for the conditional probability of zero-photon detection in a time interval Δ:

$$P_0 = \exp\Big(- n(\Delta,I) \Big), \tag{3}$$

where I is the field intensity and $n(\Delta,I)$ is the signal strength averaged over the time interval Δ. The quantum mechanical treatment (Walls & Milburn, 1994) also gives the same expression for a single mode photon field. The above expression of P_0 is valid for a signal of constant intensity. The intensity of the field, in general, does not remain constant but fluctuates. The fluctuations influence the observed probability and its dependence on signal strength. The effect of the fluctuations depends upon the time-scale of fluctuations, Δ the duration of a single event, and T the time taken in the measurement of the probability. The nature of the field determines the strength and time-scale of fluctuations. The intensity of a coherent photon field does not fluctuate at all, at least during its coherence time τ_c. The field subsequently changes its nature due to various interactions and eventually becomes a thermal field. The intensity of a thermal field fluctuates with a time-scale of τ_{th}. We shall call it thermalization time. The fluctuations of the field during this time can be described by a distribution function $\rho(I)$, which Janossy (1959) has shown is given by:

$$\rho(I) = \exp\left(- \frac{I}{I_0} \right) \frac{\mathrm{d}I}{I_0}, \tag{4}$$

where I_0 is the average intensity (Walls & Milburn, 1994), the associated amplitude being a Gaussian with zero mean value. The parameters τ_c and τ_{th} specify a photon field in the semi-classical framework, and their values determine the observed probability of zero-photon detection for any choice of Δ and T. If $\tau_{th} < \Delta$, then the field intensity fluctuates very rapidly and the fluctuations are to be taken into account in calculating the outcome of each event. One has to take a thermal average of signal intensity and signal strength in each event. The thermal averaged values I_0 and $n(\Delta,I_0)$ will be the same for all events. The fluctuations will not alter the dependence of the conditional probability on signal strength. Equation (3) will continue to describe the dependence. Similarly if $\tau_c > T$, then the signal intensity will not fluctuate during a measurement of the probability and eqn (3) will continue to describe the dependence of probability on signal strength. However, if $\tau_c > \Delta$ and $\tau_{th} < T$, then the intensity of the field can be considered unchanging during an event but fluctuating among different events. The measured probability will be an average over the intensity distribution [eqn (4)]. It is given (Walls & Milburn, 1994) by

$$P_{0T} = \frac{1}{\Big(1 + n(\Delta,I_0) \Big)}, \tag{5}$$

where $n(\Delta,I_0)$ is the thermal averaged signal strength and the subscript T in the probability is added to indicate thermal averaging. The procedure of thermal averaging becomes complicated in the semi-classical framework in the limit $I_0 \to 0$, but it leads to photon bunching in the quantum framework. Other values of τ_c and τ_{th} do not yield analytical expressions. The derivation of eqn (3) assumes a single mode radiation field, and one has to take another average over the distribution of modes. However, eqn (3) remains valid for any distribution if $\tau_c > T$ or $\tau_{th} < \Delta$ for every mode. The predictions given by eqns (3) and (5) do not differ much for stronger signals with $n(\Delta,I) > 5$ and are difficult to observe for weaker signals with $n(\Delta,I) < 0.1$. Only signals with their strength in the range

$0.1 < n(\Delta,I) < 5$ can discriminate between the two situations. It is generally believed that in the discriminatory range of signal strength, the probability in other situations will lie between the predictions given in eqns (3) and (5). Since the observed probability in biophoton signals is in excellent agreement with eqn. (3) at different values of signal strength, one does not need to consider other situations.

The value of the signal strength also depended upon a parameter Δ, which was adjustable and was varied by three orders of magnitude. The variation allowed us to obtain a signal of discriminatory strength in every portion of the delayed luminescence curve. Our measurements using discriminatory signals showed an excellent agreement with predictions of eqn. (3). The agreement implied that the fluctuations in the intensity of the field, if any, were undetectable and the average intensity over any interval Δ was the same in the entire duration T. Such a situation could occur if either $\tau_{th} < \Delta$ *or* $\tau_c > T$ for all modes of the radiation. The former condition requires that τ_{th} was less than 10 μs, the minimum value of Δ used in our measurements. If it were so, then the fluctuations in the intensity were very rapid and more than one million thermalization cycles would have spanned in 10s, the time of a measurement of the probability for the choice $T = 10$ s. A million thermalization cycles would reduce the fluctuations in the observed probability ratio. One expects the fluctuations in the probability ratio to be around 0.1%—the inverse of the square root of one million. The standard deviation of the probability ratio for coherent radiation is 2.1% for $T = 10$ s. The observed standard deviation is an order of magnitude higher than expected. Perhaps, the expectation is not based on correct assumptions. Fluctuations are not so rapid and the intensity of each event is not thermally averaged. In the absence of thermal averaging, one expects the standard deviation to be around 2.2% in a data set of 2000 repeated measurements in all cases. The standard deviation is only a few percent in different cases given in Table 1. It suggests that an individual event is not thermally averaged and τ_{th} in a biophoton signal is larger than Δ. A small τ_{th} is problematic to any classical equilibrium model (Christen, 1970) of delayed luminescence. The problem stems from a need to generate two widely different time-scales characterized by τ_{th} and τ_τ. A single chemical reaction can generate only one time-scale through its equilibrium constant. Generation of two time-scales requires two (or more) reactions and a mechanism to coordinate them in the temperature range of 15–50°C used in our measurements. We do not have any other evidence of the existence of such a mechanism. We, therefore, believe that the possibility $\tau_{th} < \Delta$ is incorrect and the coherence time of all modes emitted in delayed luminescence satisfy $\tau_c > T$. Since we used $T \leqslant 100$ ms in the decay region and $T \leqslant 10$ s in the tail region, the coherence time of all modes was more than 100 ms in the decay region and more than 10 s in the tail region.

The above estimate of the coherence time was limited by the restriction in the value of T, whose value was not possible to increase in the decay region of the signal. The continuous measurements at successive intervals diluted the effect of this restriction. The biophotons were found coherent in contiguous intervals with a coherence time longer than the interval in every measurement. One can, perhaps, infer that the coherence time was longer than the entire duration of continuous measurements by combining the results of all measurements. The continuous measurements were carried out for 200 s in the decay region and for 5 hr in the tail region. Again, the measurements in both regions had a common interval. The signal was, therefore, coherent for 5 hr. The coherent signal contained contributions of both light induced and spontaneous biophoton emissions. Consequently, biophotons were coherent in both types of emissions. Even though we have observed the coherent nature of radiation emitted only in delayed luminescence, we believe it to be a general result; the nature of biophotons is thus indeed coherent.

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