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THE REVERSIBLE DECLINE OF OXYGEN FLASH YIELDS AT HIGH FLASH ENERGIES

EVIDENCE FOR TOTAL ANNIHILATION OF EXCITATIONS IN PHOTOSYSTEM II

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The yield of oxygen from cells of *Chlorella vulgaris* illuminated for 0.5 μs with 596 nm laser light reversibly declines at flash energies much greater than those required for saturation. The decline in oxygen flash yields is specifically related to the optical cross-section per Photosystem (PS) II trap. The effect can be explained as the result of a total annihilation process which occurs either at the PS II trap (with very low probability) or in the PS II antenna (with very short lifetime). Evidence from separate experiments is discussed which suggests that the process occurs at the PS II trap. The probability of this process is about 10^{-4}.

Introduction

Since the pioneering experiments of Emerson and Arnold [1], short, bright flashes of light have been used in the investigation of photosynthesis. Recent experiments using picosecond laser pulses of high energy have demonstrated that at high excitation rates, fluorescence yields from illuminated chloroplasts and algae are greatly reduced [2,3]. The decrease in fluorescence yields at high flash energies has been attributed to interactions between excited states in the photosynthetic antenna or in the traps which result in the nonfluorescent loss of all or part of the energies of the interacting states. Effects of such interactions on the yield of photosynthetically effective photochemistry (e.g., oxygen evolution, *P*-700 photooxidation, etc.) have not been reported.

In a previous study [4], we used submicrosecond pulses of laser light to determine optical cross-sections for oxygen evolution in *Chlorella*. We observed that at flash energies much greater than those required to saturate oxygen production, flash yields were reduced in a completely reversible-manner. In this report, we provide evidence that this effect depends specifically on the absorption of light by PS II and may result from losses due to the interactions of excited states in the antenna or traps of PS II.

Materials and Methods

Axenic cultures of the green alga *Chlorella vulgaris* were grown at 20°C in continuous light as described previously [4]. Illumination levels were $2–3 \cdot 10^5$, $8 \cdot 10^3$ and $1–2 \cdot 10^2$ erg $\cdot$ cm$^{-2} \cdot$ s^{-1} for the cells grown in the high, intermediate and low high fields, respectively. Cells were collected by centrifugation and resuspended in an electrolyte buffer (0.1 M KH_2PO_4, 0.05 M NaCl, 0.05 M $NaHCO_3$, 0.01 M KCl, pH 7.8) at an equivalent chlorophyll concentration of 50–90 μM Chl ($a+b$). Chlorophyll concentrations were determined from ethanol extracts of the cells [4].

For most cross-section measurements, relative oxygen flash yields (Y_{O_2}) were determined as de-

scribed previously [4] and the following is a brief summary of the measurement protocol. Algal cells resting on the bare platinum surface of a Pickett-type oxygen polarograph were uniformly illuminated with a continuous train of short saturating 596 nm laser flashes at a rate of one flash/2 s. After oxygen flash yields had become constant (steady state) the energies of two consecutive laser flashes were varied. Laser flash energies were then returned to the preceding saturating levels and flash illumination was continued. When steady-state oxygen flash yields had been reestablished, the entire sequence could be repeated with different laser flash energies. Y_{O_2} values were calculated as the ratio of yield of the first attenuated flash to the yield of the preceding full intensity flash. In this way an entire light saturation curve could be rapidly measured on cells perturbed only briefly from steady-state conditions.

Laser flash energies were decreased from saturating (maximum oxygen flash yields) by the use of calibrated filters. Flash energies were increased from saturating levels by removing filters, increasing the total laser charge voltage or both. Increasing the laser charge voltage increased both the total laser flash energy and the flash length. The complete length of the saturating laser flashes was 450–500 ns. At the highest supersaturating flash energies used, the complete flash length had increased to 750 ns.

For the experiments in which oxygen flash yields and fluorescence yields were measured simultaneously, the protocol above was modified slightly. After steady-state oxygen flash yields had been established, five consecutive flashes were attenuated. Y_{O_2} values were then calculated as the ratio of the fifth attenuated flash yield to the steady-state yield.

The effect of laser flash energies on the relative yield of fluorescence was measured using a dim blue test flash (0.9 μs full duration at half maximum) from a xeon flashlamp (Stroboslave, Type 1539-A) triggered 30 μs after the laser flash. The light from the flashlamp was filtered with blue band-pass filters and focused into the same arm of the dual-armed fiber-optics light-pipe that was used for the laser light. Fluorescence from the algae was delivered by the second arm of the light-pipe to a gated photomultiplier system [5] blocked with a 680 nm interference filter. The photomultiplier was gated on for 1 μs at the time of the test flash. The signal-to-noise ratio from the photomultiplier monitoring the fluorescence excited by the test flash was such that five consecutive laser flashes had to be averaged for an accurate fluorescence determination. The test flash produced no detectable oxygen production by the algae.

Measurements of the effects of nanosecond duration laser flashes employed two nitrogen laser pumped rhodamine dye lasers. The protocol for these experiments was otherwise the same as that described by Mauzerall [5]. The second low-energy laser flash was delivered 30 μs after the main pulse from the first dye laser. The complete duration of the main dye laser flash was measured to be 5 ns.

The relative increase in the yield of fluorescence measured 30 μs after a laser flash ($\Delta\phi(30)$) was calculated as $1-(F_t/F_0)$ where F_t is the relative fluorescence of algae exposed to a test flash given 30 μs after the main flash and F_0 is the relative fluorescence of algae exposed to a test flash presented in the absence of a preceding laser flash. $\Delta\phi(30)$ values were subsequently normalized to a maximum yield increase of unity.

Results and Discussion

Fig. 1 shows the light-saturation behavior of the relative oxygen flash yields (Y_{O_2}) from *Chlorella*

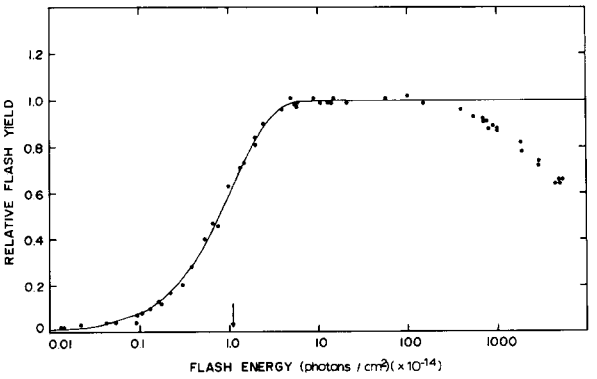

Fig. 1. Light-saturation behavior for relative oxygen flash yields from cells grown in intermediate light levels. The solid curve is the cumulative one-hit Poisson distribution (see text) calculated for $\sigma_{O_2}=90$ Å^2. The arrow indicates the flash energy corresponding to an average of one hit/PS II trap per flash. $7.1\cdot 10^{-16}$ mol Chl/cell; Chl a/Chl $b=0.33$.

cells illuminated with short flashes of 596 nm light. The total laser flash energy at the electrode surface varies by more than five orders of magnitude from about 10^{12} to about $5 \cdot 10^{17}$ photons/cm^2 per flash. As flash energies increase from their minimum values, Y_{O_2} values first increase to a maximum and then remain constant over a roughly 50-fold further increase in flash energy. At still higher (supersaturating) flash energies, Y_{O_2} values slowly decline. This light-saturation behavior is essentially identical to that described previously [4].

For flash energies less than about 10^{16} photons/cm^2 per flash, the light-saturation behavior of Y_{O_2} is well described by the 'cumulative one-hit' Poisson distribution (Fig. 1, solid curve):

$$Y_{O_2} = 1 - \exp(-\sigma_{O_2} E) \quad (1)$$

where E is the total flash energy per cm^2 at the electrode surface and σ_{O_2} is the effective optical cross-section for oxygen production by PS II. This simple light-saturation behavior will be obtained if the same photochemical effect (oxygen evolution) is produced by all PS II traps which receive the energy from one or more photons (hits) during the flash. The cross-section is a direct measure of the physical size of the target for O_2 production presented to the incoming radiation by a PS II trap and its associated pigment antenna [4]. If several PS II traps share a common antenna, σ_{O_2} is the average cross-section per trap [6]. Mauzerall [6,7] has presented evidence that about four PS II traps share a common antena.

The algal cells used to obtain the data shown in Fig. 1 were grown at an intermediate light level and contained $7.1 \cdot 10^{-16}$ mol Chl/cell. The best fit of the curve generated by Eqn. 1 to the data yields a value for σ_{O_2} of 90 Å^2. We have previously measured the 596 nm in vivo absorption cross-section for chlorophyll n *Chlorella* to be 0.29 Å^2 [4]. With this value we calculated that σ_{O_2} corresponds to an antenna size of about 310 molecules of chlorophyll per PS II trap. These values are consistent with our previous determinations for *Chlorella* cells grown under these conditions [4].

From σ_{O_2} we can calculate absorbed flash energies as the average number of hits per PS II trap per flash, $\sigma_{O_2} E$. For example, if $\sigma_{O_2} = 90$ Å^2, a flash energy of $1.11 \cdot 10^{14}$ photons/cm^2 per flash corresponds to an average of one hit/PS II trap per flash. For a 500 ns flash, this flash energy is equivalent to an average excitation rate (or absorbed flash power) of $2 \cdot 10^{-3}$ hits/PS II trap per ns.

At supersaturating flash energies, Y_{O_2} values are reduced from their maximum light-saturated values (Fig. 1). This decrease in Y_{O_2} is completely reversible on the time scale of seconds. Identical flash yields are obtained from saturating flashes given to algae before and after several hundred supersaturating flashes. Thus, PS II traps have not been irreversibly damaged or destroyed by the supersaturating flashes. However, despite illumination with flashes whose energies are in excess of 100 hits/PS II trap per flash, some PS II traps are, in effect, missed.

The extent of the reversible decrease in Y_{O_2} at supersaturating flash energies depends on the size of σ_{O_2} (Fig. 2). The data shown in Fig. 2 were obtained from cells grown in the high light (triangles) or low light (open circles) fields described above. In each case the data are the combined results of two sets of measurements using cells from different cultures. Measurements of Y_{O_2} were made over the entire flash energy range, and σ_{O_2}

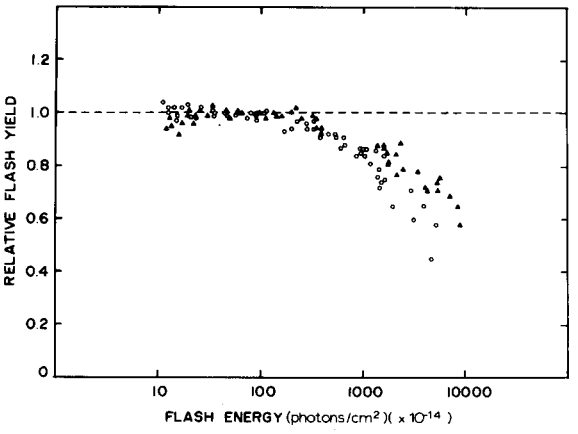

Fig. 2. The dependence of the reversible decline in relative oxygen flash yields at supersaturating flash energies on σ_{O_2}. (▲) Data from cells grown in high light; (○) data from cells grown in low light. In each case the data are the combined results of two measurements using cells grown in different cultures. High-light cells contained $1.\cdot 10^{-16}$ mol Chl/cell and had a measured σ_{O_2} of 44 Å^2. Low-light cells contained $14\cdot 10^{-16}$ mol Chl/cell and had a measured σ_{O_2} of 110 Å^2. The dashed line indicates the average maximum flash yield.

values were determined as in Fig. 1. Fig. 2 shows only the high-energy portion of these data. Despite the scatter in the data (a part of which may be due to variations in the laser flash duration), there is clearly a systematic difference between the two sets of results. Relative to the decrease in Y_{O_2} from low-light cells, the decrease in Y_{O_2} from high-light cells is displaced to greater flash energies by a factor of between 2 and 3.

Low-light cells contain nearly 9-times more chlorophyll than do high-light cells (14 vs. $1.6 \cdot 10^{-16}$ mol Chl/cell). In contrast, σ_{O_2} for low-light cells (110 Å^2) is only 2.5-times greater than σ_{O_2} for high-light cells (44 Å^2). Thus, the reversible decline in Y_{O_2} at supersaturating flash energies is not simply a function of total incident photon flux or of total cell absorption but instead depends specifically on the effective optical cross-section per PS II trap.

In Fig. 3 the data shown in Figs. 1 and 2 are replotted with flash energies recalculated as the average rate of excitation (lower scale) or the average number of hits/PS II trap per 500 ns laser flash (upper scale). Flash illumination calculated in either fashion corrects for differences in optical cross-sections as well as in laser flash lengths.

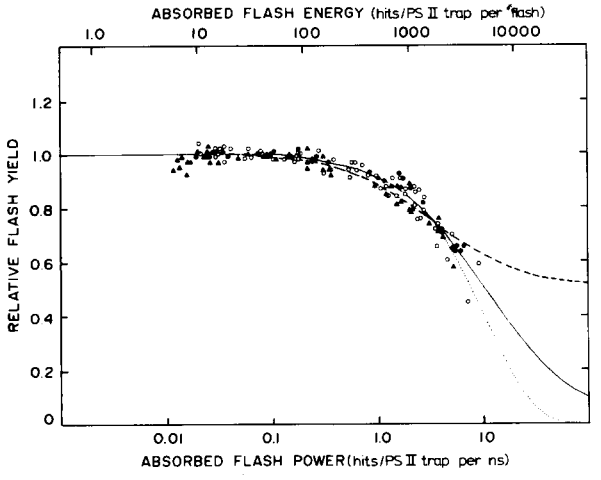

Fig. 3. The data from Figs. 1 and 2 replotted with flash energies recalculated as the average number of hits per 500 ns flash (upper scale) or as the average excitation rate (lower scale). The symbols retain their previous meanings. The dotted curve is derived from Eqn. 2 (see text) with $p = 3 \cdot 10^{-4}$. The dashed and solid curves are derived from Eqn. 3 with $\tau = 100$ ps or Eqn. 4 with $\tau = 150$ ps, respectively.

When flash illumination is so calculated, the five sets of data from the cells grown in the three light fields randomly intermingle. We conclude that the same mechanism is responsible for the reversible decline in Y_{O_2} at supersaturating flash energies in all cases and that this mechanism depends specifically on either the total photon absorption per trap or on the rate of photon absorption during a flash by a PS II trap and its associated antenna.

A decrease in Y_{O_2} which depends only on the total hits/PS II trap per flash will result if any photon absorbed at any time during the flash following an initial hit has some probability of both undoing the effect of a previous hit and inactivating the trap for the remainder of the flash, e.g., by forming a long-lived quencher. If the probability of this total annihilation with memory P is small, and the flash energies are calculated as the average number of hits per PS II trap during the flash ($X = \sigma_{O_2} E$), the relationship between Y_{O_2} and hits for large X will be (see Appendix):

$$Y_{O_2} = \exp(-pX) \tag{2}$$

A simple total annihilation process which produces no interfering products would cause PS II to act as parity detector: odd hits produce an effect, even hits do not [8]. Such behavior predicts that Y_{O_2} at very high flash energies should decline to a constant value of 50% of the saturated yields. In the absence of other effects, the flash yields at infinite flash energies will be half the saturated yields regardless of the presence or absence of 'detrapping' [7,9] at PS II traps. A semi-annihilation process is one in which two excited states interact to product a single excited state and heat. In the limiting case obtained when semi-annihilation is the only process which occurs, no decrease in Y_{O_2} will be observed at any flash energy above saturation. If this process occurs in competition with the total annihilation process, flash yields at infinite flash energy will be greater than one-half of the maximum yield. If the total annihilation process can, with some nonzero yield, also produce long-lived quencher, Y_{O_2} at high flash energies will decline to zero.

We can estimate the lifetime (τ) of the sensitive state by using the simplified kinetic approach of Mauzerall [2,7]. If a hit within the lifetime of the

sensitive state in a unit totally annihilates the state and forms a quencher of lifetime greater than the pulse length, the yield of O_2 is:

$$Y_{O_2} = \frac{T/\tau}{X + T/\tau} \tag{3}$$

If a hit within the lifetime of the sensitive state only totally annihilates the state, the yield of O_2 is:

$$Y_{O_2} = \frac{1}{2} + \frac{1}{2}\left(\frac{T/\tau}{2X + T/\tau}\right) \tag{4}$$

where T is the equivalent duration of the light pulse and τ is the lifetime of the sensitive state. It is assumed that X is large (i.e., 10 or greater). Deviations of Eqns. 2–4 are given in the Appendix. The solid and dashed curves in Fig. 3 were fitted to the data using Eqns. 3 and 4, respectively. The data appear to fit more closely by assuming that Y_{O_2} at very large flash energies is zero and not 0.5. We have not yet been able to provide flash energies large enough to test this point unambiguously. Photochemical damage at higher flash energies may also complicate the measurement.

The fits to the data shown in Fig. 3 correspond to lifetimes of about 150 ps (Eqn. 4) or about 100 ps (Eqn. 3). The scatter in the data allows fits using a 50% variation in τ about these values. If four PS II traps share a common antenna as Mauzerall [6,7] has concluded, the rate of excitation of this larger unit is 4-times greater than the rate per PS II trap. In this case, the curves in Fig. 3 correspond to lifetimes of 40 (Eqn. 4) or 25 (Eqn. 3) ps. The fluorescence lifetime in dark-adapted algae has been measured to be about 450 ps [3]. A value for τ which is very much smaller than the fluorescence lifetime would suggest that absorbed light energy can be rapidly trapped by PS II traps but that the process of detrapping [9], the escape of excitation energy from closed PS II traps, is an important component in the 450 ps lifetime of fluorescence. τ would correspond to the time an excited state spends in the PS II trap antenna before first encountering PS II traps.

The mechanism for the observed reversible decline in Y_{O_2} described by Eqn. 2 depends only on the total number of hits per PS II trap during the flash. In contrast, mechanisms described by Eqns. 3 and 4 depend on the rate of excitation. Thus, the two types of mechanisms can be experimentally distinguished by using two sets of measurements, each having flashes of different duration. For example, the data shown in Fig. 3 can be fitted with values for p of about $3 \cdot 10^{-4}$ (Eqn. 2) or with values for τ of about 100 ps (Eqn. 3 or 4). We can calculate that a 7 ns flash whose energy corresponds to 50 hits/PS II trap per flash will have a value for Y_{O_2} close to unity if the decline in Y_{O_2} is due to a low-probability event (Eqn. 2). On the other hand, if Y_{O_2} depends on the rate of excitation, we calculated a flash yield of about 0.6 (Eqn. 3) or 0.7 (Eqn. 4). A more extreme case would be obtained using picosecond flashes.

The relative yield of fluorescence from *Chlorella* cells illuminated with a short saturating flash of light is at its maximum 30 μs following the flash [10]. Fig. 4 shows the results of an experiment to determine the effect of laser flash energy on both Y_{O_2} and the relative fluorescence yield increase measured 30 μs after the laser flash ($\Delta\phi(30)$). In this experiment, technical reasons required that we average five consecutive laser flashes (see Materials and Methods). For subsaturating flash energies, Y_{O_2} values of the fifth attenuated flash are lower than those of the first attenuated flash. The dotted curve in Fig. 4 is the best fit of Eqn. 1 to the data for Y_{O_2} of the first attenuated flash. (These data are presented as Fig. 1.) At low flash energies,

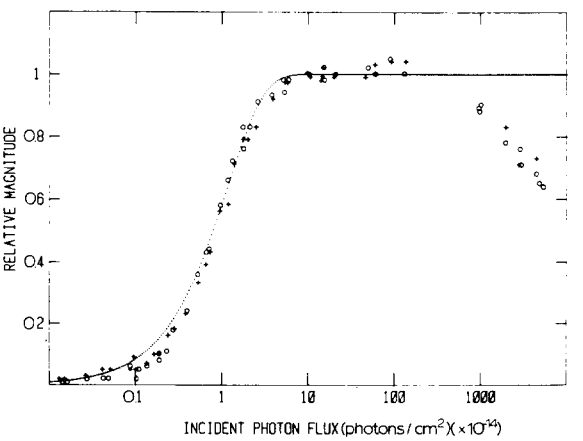

Fig. 4. Comparison of the light-saturation behavior of Y_{O_2} and $\Delta\phi(30)$. Y_{O_2} of the fifth attenuated flash (○) and $\Delta\phi(30)$ (+) were measured as described in the text. The dotted curve was calculated as the best fit to Y_{O_2} of the first attenuated flash.

Y_{O_2} values of the fifth attenuated flash fall below the dotted curve. Lowered O_2 flash yields are an expected consequence of the flash frequency and the kinetics of the O_2-producing system [4].

The data shown in Fig. 4 demonstrate that in *Chlorella* the flash energy dependences of both Y_{O_2} and $\Delta\phi(30)$ are quantitatively identical. Y_{O_2} (open circles) and $\Delta\phi(30)$ (crosses) intermingle at all flash energies tested and both show the same reversible decline at supersaturating flash energies. We conclude that in *Chlorella* the two parameters reflect the same photochemical reaction of PS II traps.

Mauzerall [2] has measured $\Delta\phi(30)$ as a function of the energy of a 7 ns flash of 337 nm light from a nitrogen laser. These results are reproduced (crosses) in Fig. 5. We have repeated these measurements using 5-ns flashes of 570 nm light from a dye laser pumped by a nitrogen laser and the results are presented as the closed circles in Fig. 5. Both sets of data are plotted versus total hits/PS II trap per flash (determined from the best fit of Eqn. 1 to the data). The solid curves labeled A, B and C were calculated from Eqn. 1 and either Eqn. 2, 4, or 3, respectively. To facilitate comparison, curves B and C were normalized to a maximum $\Delta\phi(30)$ of unity and their positions along the x-axis were shifted slightly to match curve A.

It is clear that the data shown in Fig. 5 do not show the decline in yields at the higher flash energies which we predict from Eqn. 3 or 4. Thus, we conclude that the reversible decrease in Y_{O_2} seen at supersaturating flash energies results from a low-probability event occurring during a flash in which a photon absorbed following an initial hit on a PS II trap both undoes the effect of the initial hit and also inactivates the trap for the remainder of the flash. The probability of this event is about 10^{-4}.

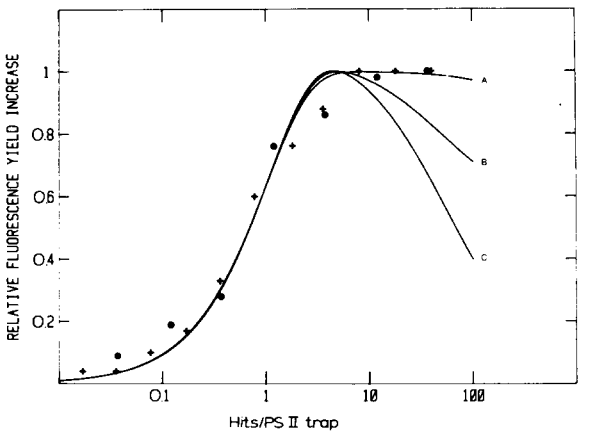

Fig. 5. The effect of nanosecond laser flash energies on $\Delta\phi(30)$. (●) Data obtained using 5-ns 570-nm dye laser flashes. (+) The data of Mauzerall [2] using 7-ns 337-nm nitrogen laser flashes. The solid curves A–C were calculated as described in the text.

The identity of the quencher in the above process is not clear. The reversible decline in Y_{O_2} is quantitatively different from the decrease in fluorescence quantum yields seen during high-energy submicrosecond laser flashes [2,3,11–14]. The observed decrease in Y_{O_2} requires that, without exception, all quanta available to a PS II trap during the entire flash are lost to oxygenic photochemistry. Lowered fluorescence quantum yields during short flashes of light have been attributed to nonradiative interactions between the excited states formed during the flash [2,3] or their dissipation by long-lived flash-generated quenchers [2,3,11–14]. The triplet state of carotenoids is believed to be the major fluorescence quencher for flashes lasting longer than a few nanoseconds [11–14]. Kramer and Mathis [15] have measured a 12 ns rise time for the formation of this state in chloroplasts. This time is considerably slower than the speed with which light energy can be trapped by PS II traps, since minimum fluorescence decay times for dark-adapted *Chlorella* cells are about 0.5 ns [3]. It thus seems unlikely that the carotenoid triplet states involved in fluorescence quenching are also responsible for the reversible decline in Y_{O_2} we report here. It is possible that chlorophyll triplet states are involved in this quenching. In any case, the key point with 'total annihilation' is that no result useful to PS II traps is available after this process.

Appendix

Statistical calculation of yields

If one excitation always remains after n hits, i.e., semi-annihilation, then the yield of O_2 is simply the cumulative Poisson distribution:

$$Y_{O_2} = P_1 + P_2 + P_3 \ldots = 1 - P_0 = 1 - e^{-x} \qquad \text{(A1)}$$

where P_n are the individual Poisson distributions and x is the average number of hits.

If there is a probablity, p, at each hit following the first that the excitations are annihilated and the trap quenched for the duration of the flash,

$$Y_{O_2} = P_1 + P_2(1-p) + P_3(1-p)^2 + \ldots = \sum_1^\infty q^{n-1} P_n$$

$$= \sum_{n=1}^{\infty} \frac{q^{n-1} x^n e^{-x}}{n!} = \frac{e^{-x}}{q} \sum_{n=1}^{\infty} \frac{(qx)^n}{n!}$$

$$= \frac{e^{-x}}{q}(e^{qx} - 1) = \frac{e^{(q-1)x} - e^{-x}}{q} = \frac{e^{-px} - e^{-x}}{1-p} \qquad \text{(A2)}$$

For small p and large x, this reduces to Eqn. 2 in the text.

If total annihilation occurs only on reexcitation within the lifetime τ of a previous excitation, and b is the probability of being in this special state (see Refs. 2 and 7):

$$b = \frac{n}{n + T/\tau} \qquad \text{(A3)}$$

where n is the number of hits to a specific unit during the equivalent pulse duration T, then the yield is:

$$Y_{O_2} = P_1 + P_2(1-b) + P_3(1-b) + \ldots$$

$$= \sum_{n=1}^{\infty} P_n - \sum_{n=2}^{\infty} P_n \frac{n}{n + T/\tau}$$

$$\approx 1 - e^{-x} - \frac{x}{x + T/\tau} \qquad \text{(A4)}$$

The Poisson weighted probability b is approximated very closely by the same saturation form with average x. This is particularly so for large x when Eqn. A4 reduces to Eqn. 3 of the text.

If total annihilation occurs on reexcitation within the lifetime τ of a previous excitation, but the trap is left open to further excitations, the yield is:

$$Y_{O_2} = P_1 + P_2(1-b) + P_3 + P_4(1-b) + \ldots$$

$$= \sum_{n=0}^{\infty} P_{2n+1} + \sum_{n=1}^{\infty} (1-b) P_{2n}$$

$$\approx \frac{1 - e^{-2x}}{2} + \frac{1 + e^{-2x} - 2e^{-x}}{2} \cdot \frac{T/\tau}{2x + T/\tau} \qquad \text{(A5)}$$

For $b = 0$, i.e., $\tau = 0$, Eqn. A1 is recovered as expected, and for large x, Eqn. A5 reduces to Eqn. 4 of the text.

Acknowledgements

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