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LETTER TO THE EDITOR

Antibunching: more than one kind in two-photon absorption

G P Hildred and A G Hall

Physics Department, University of Hull, HU6 7RX, England

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Abstract. Properties of the second moment of the photon distribution and intensity autocorrelation function are examined in a system undergoing only two-photon transitions. These properties are interpreted in terms of two kinds of antibunching, strict and temporal. It is shown that antibunching is a richer class of phenomena than usually supposed and that temporal antibunching may be easier to observe than the strict antibunching usually considered.

Bunching and antibunching are commonly defined with respect to factorisation of the second moment of the photon distribution. The criterion for antibunching being

$$\langle n(n-1)\rangle < \langle n\rangle^2 \tag{1}$$

where angular brackets denote averages. This criterion arises from notions of coherence and will here be called strict antibunching. In strict antibunching two photons are less likely to be found at the same time and place than in a random distribution. Most detectors detect a single photon, with recovery time making direct measurement of the second moment difficult. A more appropriate criterion is given by the intensity autocorrelation function: for antibunching

$$G^{(2)}(t) < \langle n(0)\rangle\langle n(t)\rangle. \tag{2}$$

This is a measure of whether another photon is more or less likely to be found at a time t after one photon has already been found. This interpretation of the physical idea of antibunching has a wider range of practical applicability than the strict criterion. It will here be called temporal antibunching.

The photon distributions of many systems are expected to be such that

$$G^{(2)}(\infty) = \langle n(0)\rangle\langle n(\infty)\rangle \tag{3}$$

and for such systems criterion (1) implies (2) and vice versa for at least the lower range of $t(t\to 0)$. However it will be shown that for a simple two-photon absorbing system equation (3) is not satisfied, and indeed it is possible to have

$$G^{(2)}(\infty) < \langle n(0)\rangle\langle n(\infty)\rangle. \tag{4}$$

The totally relaxed $G^{(2)}(\infty)$ is largely independent of the starting conditions, and in such cases the temporal antibunching condition (2) becomes independent of the strict condition (1) for larger values of t. This indicates that the two kinds of antibunching are distinguishable. These two kinds of antibunching are explored in a two-photon absorbing system.

Simaan and Loudon (1975) have made a study of a system in which only two-photon transitions are allowed. In such systems the photon probability distribution P_n splits into the two series given by n odd and n even, which are totally independent except for the normalisation condition:

$$\sum_{n\ \mathrm{even}} P_n + \sum_{n\ \mathrm{odd}} P_n = 1. \tag{5}$$

Hence it is possible to write

$$\sum\nolimits^{\mathrm{e}} P_n = 1 - K, \qquad \sum\nolimits^{\mathrm{o}} P_n = K, \qquad 0 \leqslant K \leqslant 1 \tag{6}$$

where e and o are used to denote summations over even and odd n respectively. K is conserved by the system and therefore depends only on the initial photon distribution. The steady state photon distribution can be written

$$P_n = \begin{cases} r^{n/2}(1-r)(1-K) & n \text{ even} \\ r^{(n-1)/2}(1-r)K & n \text{ odd} \end{cases} \qquad 0 \leqslant r \leqslant 1 \tag{7}$$

where r is the ratio of the number of atoms in the upper state to the number in the lower state. Then in the steady state

$$\begin{aligned} \bar{n}_{\mathrm{e}} &= \sum\nolimits^{\mathrm{e}} nP_n = \frac{2(1-K)r}{1-r} \\ \bar{n}_{\mathrm{o}} &= \sum\nolimits^{\mathrm{o}} nP_n = \frac{K(1+r)}{1-r} \end{aligned} \tag{8}$$

$$\langle n \rangle = \bar{n}^{\mathrm{e}} + \bar{n}^{\mathrm{o}} \tag{9}$$

$$\langle n(n-1) \rangle = \frac{2r}{(1-r)^2}[(1+r) + 2(K - Kr + r)]. \tag{10}$$

The criterion for strict antibunching is examined below.

The intensity autocorrelation function can be written (Jakeman and Pike 1971, Hildred and Hall 1978)

$$G^{(2)}(t) = \sum_{m,n} nmP_m(0)P_n(m-1|t) \tag{11}$$

where $P_n(m-1|t)$ is the conditional probability of finding an $|n\rangle$ photon state at a time t after a photon has been detected in the pure $|m\rangle$ photon state. It satisfies the initial condition

$$P_n(m|0) = \delta_{mn}. \tag{12}$$

In the system with only two-photon transitions, because there is no connection between even and odd series, and because if m is even $m-1$ is odd and vice versa,

$$\begin{aligned} G^{(2)}(t) &= \sum_m\nolimits^{\mathrm{o}} mP_m(0) \sum_n\nolimits^{\mathrm{e}} nP_n(m-1|t) + \sum_m\nolimits^{\mathrm{e}} mP_m(0) \sum_n\nolimits^{\mathrm{o}} nP_n(m-1|t) \\ &= \sum_m\nolimits^{\mathrm{o}} mP_m(0)\bar{n}^{\mathrm{e}}(m-1|t) + \sum_m\nolimits^{\mathrm{e}} mP_m(0)\bar{n}^{\mathrm{o}}(m-1|t). \end{aligned} \tag{13}$$

In the latter form of e.g. (13) conditional first moments have been introduced in a manner analogous to equations (8). These satisfy equations like those for unconditional $\bar{n}^{e,o}$ except for initial conditions which are determined by equation (12) to be

$$\bar{n}^{e,o}(m|0) = m. \tag{14}$$

$\bar{n}^{e}(m|t)$ therefore corresponds to $K=0$ and $\bar{n}^{o}(m|t)$ to $K=1$. The long time behaviour $t\to\infty$ is independent of the precise initial state and tends to the same limits as those for unconditional $\bar{n}^{e,o}$. Using equations (8) with the appropriate values for K

$$\bar{n}^{e}(m-1|\infty) = \frac{2r}{1-r}, \qquad \bar{n}^{o}(m-1|\infty) = \frac{1+r}{1-r}. \tag{15}$$

Then

$$G^{(2)}(\infty) = \bar{n}^{o}(0)\bar{n}^{e}(\infty, K=0) + \bar{n}^{e}(0)\bar{n}^{0}(\infty, K=1) \tag{16}$$

and in the steady state

$$G^{(2)}(\infty) = \langle n\rangle^2 - K\langle n\rangle + \bar{n}^{e} = \frac{2r}{(1-r)^2}(1+r). \tag{17}$$

It is notable in equations (16) and (17) that $G^2(\infty)$ does not satisfy the factorisation (3); and indeed in the steady state does not depend on K, in contrast to $\langle n(n-1)\rangle$.

Using the above results in the steady state the criterion for strict antibunching is

$$K > [2r(1+r)]^{1/2}(1-r)^{-1} \tag{18}$$

and for temporal antibunching is

$$K > \{[2r(1+r)]^{1/2} - 2r\}(1-r)^{-1}. \tag{19}$$

Values of K and r for antibunching fall to the right and below the limiting curves plotted in the Kr plane in figure 1. There is an absolute limit ($\sqrt{5}-2$) to the value of r

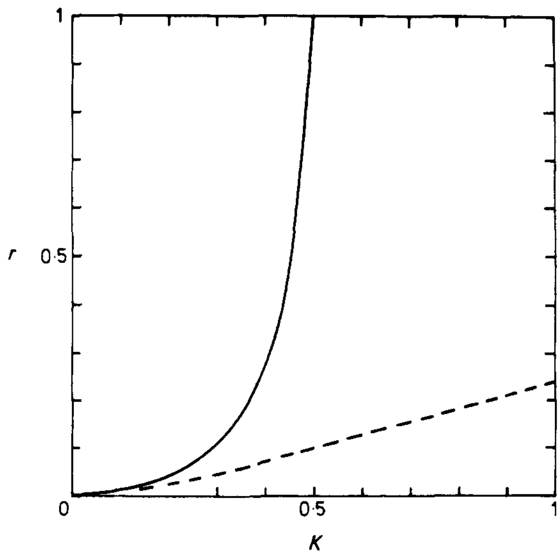

Figure 1.

for which strict antibunching can occur, with no such restriction on temporal antibunching. The region for temporal antibunching is far greater than, and contains that, for strict antibunching. Therefore temporal antibunching will be observed in the steady state whenever strict antibunching occurs, but may also be observed when strict antibuncing is not present. Conditions for temporal antibunching are easier to satisfy.

Although antibunching in two-photon absorbing systems may be observed in $G^{(2)}(t)$ is appears in a disguised form. In a system satisfying the factorisation (3) and strict antibunching the steady state $G^{(2)}(t)$ will start at $t=0$ with a value less than $\langle n\rangle^2$ and eventually rise to this value as $t\to\infty$. However comparing equations (10) and (17) it is seen that in the steady state of the two-photon absorbing system $G^{(2)}(0)$ is always greater than $G^{(2)}(\infty)$ by a factor $2(K-Kr+r)$ which is always positive $0<K, r<1$. $G^{(2)}(t)$ will always fall to its limiting value as $t\to\infty$, even if strict antibunching occurs. Thus the antibunching $G^{(2)}(t)$ has a shape more likely to be associated with bunching.

The above discussion depends on the strict separation of the photon probability distribution into even and odd series. This only occurs when the system is strictly limited to two-photon transitions. If the effect of other transitions proceeds at a rate greater than that of two-photon transitions then temporal antibunching will be masked even if strict antibunching occurs. If the effect of other transitions can be kept to a very slow rate then $G^{(2)}(t)$ will show a fall to temporal antibunching followed by a slow rise to factorisation.

It is evident from study of the two-photon absorbing system that photon antibunching is a richer phenomenon than is usually supposed. This is especially the case in systems for which events separated by large times do not satisfy the ergodic factorisation (3). In absence of this factorisation it is possible to define strict antibunching and temporal antibunching, both of which may be found in two-photon absorbing systems.

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