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LETTER TO THE EDITOR

The production of photon antibunching by two-photon absorption

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Abstract. The two-photon absorption process is, in principle, capable of producing photon beams with second-order coherence less than unity. The dependence of the observability of such values on the initial state of the beam, the absorption path length and the detector sensitivity is outlined. It is concluded that the main problem is not the low intensity involved but rather the low two-photon absorption cross section.

The production of photon beams with second-order coherence different from the values of 1 and 2 which correspond to coherent and chaotic states has been the subject of much interest recently (Stoler 1974, McNeil and Walls 1975, Simaan and Loudon 1975). Values of less than unity for the second-order coherence function (at $\Delta x = 0$), $g^{(2)}(0)$, cannot be obtained with a classical electromagnetic field as this would correspond to a negative variance of the field intensity. Beams with $g^{(2)}(0) < 1$ are sometimes visualized in terms of the 'antibunching' of particle-like photons. Photon counting statistics which are not capable of being predicted in terms of the semiclassical theory have been observed only extremely rarely (Clauser 1974) and for this reason alone the observation of a value of $g^{(2)}(0)$ less than unity would be of the greatest interest. McNeil and Walls (1975) have suggested the production of beams with $g^{(2)}(0) > 2$. Such values of $g^{(2)}(0)$ are not inconsistent with the classical treatment of the photon field.

The development with time of the quantum statistics of a photon beam undergoing two-photon absorption has been analysed by Simaan and Loudon (1975) and McNeil and Walls (1974). The present author has also independently calculated the development using a form of numerical integration applied to an equation of the form of equation (7) of Simaan and Loudon's paper with N_2 set to zero. The main purpose of this letter is to analyse the possibilities for the experimental observation of the antibunching effect. Simaan and Loudon note that the effect would be difficult to observe since $g^{(2)}(0)$ values significantly less than unity occur only when the mean number of photons in the beam is comparable with unity. However, the relevant parameter is not the total number of photons in the beam but rather the number of photons which are stimulating the detector at any instant. Thus if the 'photons' as detected by the detector are localized (that is, the observation of a count affects the field only in a localized region) then in principle a given deviation of $g^{(2)}(0)$ below unity could correspond to an arbitrarily high intensity if the localization length l were small enough. In practice the localization length is likely to be of the order of 0·1 m and in this case $\bar{n} = 1$ corresponds to a flux of 3×10^9 photons s^{-1} or approximately 10^{-9} W. Although this would not appear to limit the observation of the antibunching effect too severely there are more serious problems related to the low value of the cross section of the two-photon absorption process.

The cross section of the two-photon absorption process is proportional to the intensity of the photon beam. Thus the absorption coefficient $(dI/dx)/I$ can be enhanced arbitrarily by increasing the power in the incident beam. However, increasing the intensity does not greatly affect the rate of change of $g^{(2)}(0)$. In fact, for a coherent beam, an increase in intensity leads to a slower change in $g^{(2)}(0)$. The dependence of $g^{(2)}(0)$ on $\bar{n}$ and the distance of propagation (x m) through the absorption medium, is shown in figure 1. The notation used is that of Simaan and Loudon (1975) but the values were

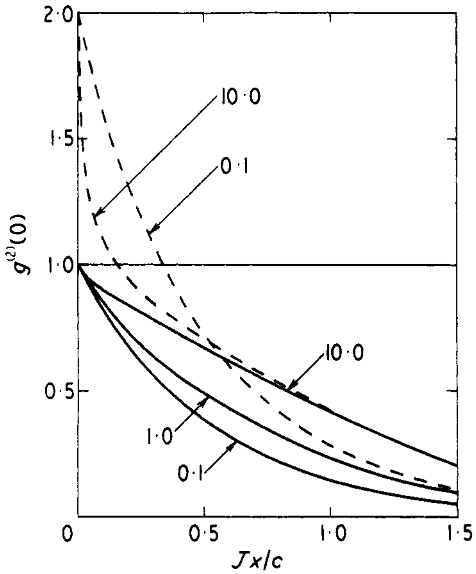

Figure 1. A graph of the degree of second-order coherence, $g^{(2)}(0)$, of a photon beam which has undergone two-photon absorption over a path length of x m, plotted as a function of Jx/c for several values of $\bar{n}$ (shown on figure). The full lines correspond to fully coherent states and the broken lines to chaotic photon states.

obtained by a numerical integration. These results appear to suggest that a very intense chaotic or a very weak coherent beam should be used to maximize the deviation of $g^{(2)}(0)$ from unity. There is, however, another important consideration which must be taken into account. To obtain statistics which are good enough to justify a claim that a value of $g^{(2)}(0)$ less than unity has been observed, one requires a large flux of photons. If the sensitivity of the coincidence detector is S and the intensity I is given by $\alpha\bar{n}$, so that the mean number of coincidences in a time interval of length t is $\alpha^2 St\langle n(n-1)\rangle$, then the fractional standard deviation of the observed coincidence rate in a given channel is $(\alpha^2 St\langle n(n-1)\rangle)^{-1/2}$. This must be less than the actual deviation of $g^{(2)}(0)$ from unity if the antibunching effect is to be observed. That is,

$$\alpha^2 St > \left[\langle n(n-1)\rangle\left(1-\frac{\langle n(n-1)\rangle}{\langle n\rangle}\right)^2\right]^{-1} \equiv \alpha^2 St_0.$$

For small values of Jx/c it can be shown that

$$\alpha^2 St_0 = c^2/4(Jx\bar{n})^2 + c/Jx\bar{n} + c/2Jx\bar{n}^2 + O[(Jx/c)^0].$$

Figure 2 shows the value of $\alpha^2 S t_0$ as it deviates from this relation for higher values of Jx/c. The curves possess minima which correspond to the ideal compromise between the increase in the antibunching effect and the decrease in intensity which result from an increase in the path length in the absorption medium.

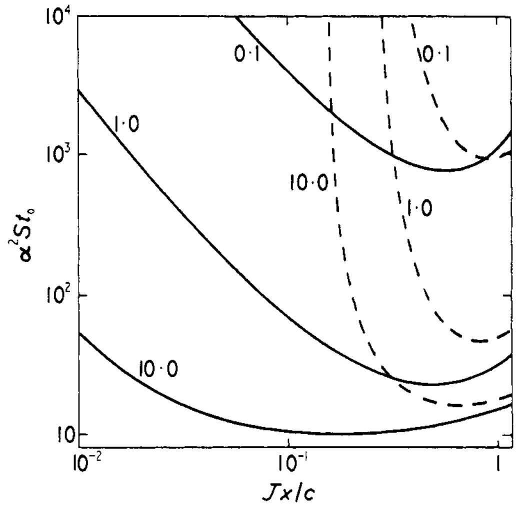

Figure 2. A log–log plot of the parameter $\alpha^2 S t_0$ against Jx/c for several values of $\bar{n}$ (shown on figure). The time interval t_0 s is the time required to obtain sufficient statistics to observe a deviation in the degree of second-order coherence below unity greater than that corresponding to one standard deviation in the coincidence rate. The full lines correspond to fully coherent states and the broken lines to chaotic photon states.

The scale of the horizontal axis, (c/J), of these graphs is determined by several parameters including the cross section of the absorption process. The experimental evidence (Gold 1969) is that $P/F^2 \equiv \beta$ is about 10^{-58} m^4 s for a non-resonant process and about 10^{-50} m^4 s for a resonant process (P is the probability per second of a two-photon absorption event by a single atom in a photon flux of F photons s^{-1} m^{-2}). Thus $c/J = lA/Nc\beta$ m, where A(m^2) is the cross sectional area of the photon beam, N(m^{-3}) is the number of atoms per unit volume and c m s^{-1} is the speed of light. For reasonable values of l, A, N and β this length is very large. For example $Jx/c = 0{\cdot}1$, $l = 1$, $A = 10^{-6}$, $N = 10^{29}$, $\beta = 10^{-50}$ correspond to a path length in the absorption medium of 3×10^4 m. Because measurements over such path lengths are almost certainly impractical, any experiment is likely to have $Jx/c \ll 1$. In this case, for a coherent beam,

$$\alpha^2 S t_0 \simeq (c/J)^2[1/4(x\bar{n})^2]$$

and the condition for the observability of the antibunching effect can be re-expressed as requiring that the number of coincidences observed in the region of the antibunching dip must be greater than

$$\left(\frac{c}{2Jx}\right)^2 = \left(\frac{lA}{2Nc\beta x}\right)^2.$$

With the above data and a path length of 10 m, $1{\cdot}5 \times 10^4$ coincidences would have to be counted in the region of the dip to make the standard deviation as small as the amount by which $g^{(2)}(0)$ is less than unity.

Thus the limiting feature is not necessarily the low intensity required to produce a significant antibunching effect. For realistic path lengths the improvement in statistics obtained by increasing the intensity more than outweighs the decrease in the deviation of $g^{(2)}(0)$ from unity. To maximize the observability of the effect, the system should be designed to minimize the cross sectional area and the localization length of the photon beam, and to maximize the cross section constant β and the atomic density as well as the photon intensity. The effect of unwanted one-photon absorption would not be a problem (except that in extreme cases it might reduce the effective path length and the final intensity) as such processes have no effect on the second-order coherence function.

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