Photon antibunching

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A review is given of recent theoretical studies devoted to the problem of generating radiation fields that exhibit the opposite of the well-known bunching of photons observed in light from thermal sources, the so-called antibunching effect. It is made clear that this phenomenon reflects the corpuscular nature of light and, hence, cannot be interpreted in terms of classical electrodynamics, needing, instead, the quantum-mechanical formalism for its description. It is shown in some detail that nonlinear interaction mechanisms like multiphoton absorption and parametric three-wave interaction are suited to change the photon statistical properties of incident (in most cases coherent) light such that the output field will be endowed with antibunching properties. Special emphasis is given to the problem of correctly specifying the dimensions of the mode volume occurring in the usual single-mode treatment of the field, which is, in fact, of great practical interest, since the magnitude of the antibunching effect is determined by the inverse average number of photons contained in that volume. In a later section it is pointed out that destructive interference with a coherent reference beam provides a means of (a) effectively enhancing photon antibunching that is already present in a high-intensity field, through reduction of the intensity, and (b) transforming phase fluctuations produced in a Kerr medium into antibunching-type intensity fluctuations. On the other hand, there exists a way of directly generating light with antibunching properties, the physical mechanism being resonance fluorescence from a single atom. The main features of this technique, both theoretical and experimental, are outlined, including a discussion of the first experimental results obtained a few years ago.

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I. INTRODUCTION

Without exaggeration, one can say that the advent of fast photoelectric detectors opened a new area of experimental optical research by making fluctuation phenomena occurring in optical fields accessible to observation. The pioneering work in this field has been done by R. Hanbury Brown and R. Q. Twiss (1956), who for the first time measured intensity correlations in a light beam originating from a thermal source, thus stimulating a good deal not only of experimental but also of theoretical progress in what is nowadays known as quantum optics.

By the way, it is interesting to note that the investigations of Brown and Twiss were motivated by a rather practical need, their goal being an improved version of Michelson's stellar interferometer, aimed at the elimination of the disturbing influence of atmospheric scintillations. While in Michelson's device light from a star is impinging on two distant mirrors and the reflected beams are made to interfere [for details see, e.g., Mandel and Wolf (1965)], Brown and Twiss proposed to focus the incident light, by means of two large reflectors, onto two photomultiplier tubes and to measure correlations in the output currents of the detectors as a physical equivalent to the fringe visibility in Michelson's interferometer. In fact, the laboratory experiment of Brown and Twiss (1956) mentioned above, which contributed so much to an understanding of the quantum features of radiation, was primarily intended to serve as a demonstration of the feasibility of the novel astronomical observation technique they had in mind.

Later on, photodetectors counting single photons were also employed in the study of the Brown and Twiss effect, and not only spatial but also temporal intensity correlations became a subject of investigations. Let us briefly describe a typical experimental device which may be viewed as the prototype of modern photon counting techniques. A quasimonochromatic light beam from a thermal source is divided by a beam splitter into two mutually coherent parts, each of which is directed to a separate detector (see Fig. 1). What is measured is delayed coincidences in the counting rates for both detectors, i.e., those events are recorded when the first detector counts a photon and the second detector does so τ seconds later.

The experiment reveals the following feature characteristic of thermal light: The coincidence counting rate, as a function of the delay time τ, exhibits a distinct peak at $\tau=0$, as shown in Fig. 2. This result indicates a tendency of the photons to arrive in pairs; hence the phenomenon has been termed "photon bunching." Since the coincidences at $\tau=\infty$ will have a purely random character, it follows from Fig. 2 that the (nondelayed) coincidences are in excess of the random ones. It should be emphasized that the bunching effect is by no means peculiar from the viewpoint of classical electrodynamics—in fact, it reflects nothing else than the intensity fluctuations that are normally present in light fields, especially in those emitted by thermal sources.

The invention of the laser, which made it possible to generate radiation fields with almost fantastic properties, also stimulated renewed interest in photon statistical studies. Viewed from the theoretical standpoint, the most striking feature of laser light is its amplitude stability (provided the laser is operated in a single-mode regime not too close to threshold). In this respect, laser radiation differs from thermal light not only quantitatively, but in principle. This characteristic property of laser radiation is revealed in a Brown-and-Twiss—type experiment which now yields a coincidence counting rate practically not depending on the delay time. This means the photon bunching effect has disappeared. Moreover, one may observe the bunching phenomenon to decrease more and more when, starting from laser operation below threshold, one makes the laser pass the threshold by continuously increasing the pump power.

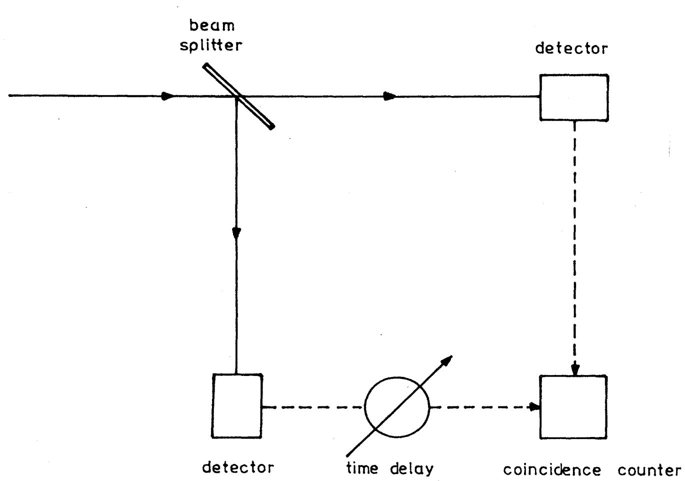

FIG. 1. Brown-and-Twiss—type arrangement for the observation of intensity correlations.

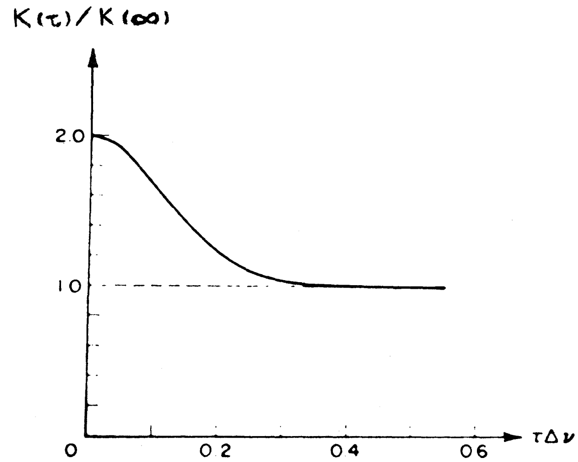

FIG. 2. The coincidence counting rate $K(\tau)$, relative to the random coincidence counting rate $K(\infty)$, vs delay time τ for thermal light of rms width $\Delta\nu$. [Theoretical curve, after Mandel (1963).]

One may ask whether those photon statistical properties exhaust all the possibilities offered, at least in principle, by nature. Specifically, the question is: May there exist an effect opposite to the bunching phenomenon which may be characterized as a deficit, rather than an excess, of (nondelayed) coincidences with respect to the random ones? In the framework of quantum electrodynamics, the answer is readily given, and it is confirmative. In fact, a lot of quantum-mechanical states for the radiation field can be specified which would exhibit such an "antibunching" effect. In particular, the well-known energy eigenstates for a single-mode field (Fock states), corresponding to definite values of the photon number, are of this type. The essential physical question, however, is how to produce such peculiar states of the field in practice, or at least in a Gedankenexperiment. This problem is of fundamental interest, since antibunching has no analog in classical optics. Hence, this effect shows up the intrinsic quantum nature of the radiation field.

The aim of the present article is to review the main features of recent theoretical studies devoted to the problem of generating electromagnetic fields with antibunching properties. Moreover, the feasibility of such experiments will be discussed in some detail, and a brief account will be given of the first experimental efforts to demonstrate the antibunching effect.

II. PHYSICAL AND FORMAL ASPECTS OF PHOTON BUNCHING AND ANTIBUNCHING

A. Intensity fluctuations

Before getting absorbed in the quantum mechanical formalism needed for a correct description of the antibunching phenomenon, I shall look at a simplified approach to photon statistics based on the classical concept of light waves and, alternatively, on a naive photon picture. It will provide some insight in the physical essence of antibunching, and, moreover, suggest possible experimental ways of producing light beams displaying this phenomenon.

Basic to the description of photocounting in the wave theory of light is the well-established fact that a photodetector responds to the instantaneous intensity $I(t)$ of the optical field acting on the detector surface. More precisely speaking, since a finite response time τ_{resp} must be ascribed to any detector, the probability of counting a photon during a time interval of length τ_{resp} is proportional to the integral of $I(t)$ over that interval. Assuming τ_{resp} to be short compared to the period characteristic of the duration of individual intensity fluctuations (in case of thermal light, this time scale is given, by order of magnitude, by the coherence time, i.e., the coherence length divided by the velocity of light), we may neglect the difference between the instantaneous intensity and its time average over the response time of the detector.

Let us first consider a stationary (however randomly fluctuating!) field. What can be immediately measured in this case is the time-averaged counting rate for a photodetector

$$R=s\lim_{T\to\infty}\frac{1}{T}\int_{-T/2}^{T/2}I(t)dt\equiv s\bar{I}\ , \tag{2.1}$$

where s characterizes the detector sensitivity. According to the well-known ergodic theorem, the time-averaging procedure indicated in Eq. (2.1) yields the same result as averaging over the statistical ensemble representative of the field.

It should be noted that the present analysis is easily extended to fields which are periodic in time. Then, the averages are to be taken over a period.

The experimental setup described in Sec. I (see Fig. 1) allows one to measure coincidence counting rates for two detectors. Since the effect of a beam splitter is to divide the incident beam into two parts with half the (instantaneous!) intensity, respectively, of the original beam, $I(t)$, we find the coincidence counting rate to be

$$K(\tau)=\frac{s^2}{4}\tau_{\text{resp}}\lim_{T\to\infty}\frac{1}{T}\int_{-T/2}^{T/2}I(t)I(t+\tau)dt$$

$$\equiv\frac{s^2}{4}\tau_{\text{resp}}\overline{I(t)I(t+\tau)}\ , \tag{2.2}$$

where τ denotes the delay time. [Since $K(\tau)$ is an even function of τ, we shall confine τ to non-negative values in the following.]

Now, it is well known in statistical communication theory that the autocorrelation function $\overline{I(t)I(t+\tau)}$ has an absolute maximum at $\tau=0$; this means the coincidence counting rate $K(\tau)$ quite generally obeys the inequality

$$K(0)\geq K(\tau)\quad\text{for all } \tau\ . \tag{2.3}$$

In fact, this statement immediately follows from the simple relation $0\leq[I(t)-I(t+\tau)]^2$ by time averaging [cf., e.g., Middleton (1960)].

Separating the instantaneous intensity in its mean value $\bar{I}$ and a fluctuating part $i(t)$

$$I(t)=\bar{I}+i(t) \tag{2.4}$$

and observing that $\overline{i(t)}=0$, we may rewrite Eq. (2.2) in the form

$$K(\tau)=\frac{s^2}{4}\tau_{\text{resp}}[\bar{I}^2+\overline{i(t)i(t+\tau)}]\ , \tag{2.5}$$

from which it follows that

$$K(0)=\frac{s^2}{4}\tau_{\text{resp}}(\bar{I}^2+\bar{i}^2)\geq\frac{s^2}{4}\tau_{\text{resp}}\bar{I}^2$$

$$=R'^2\tau_{\text{resp}}\ , \tag{2.6}$$

where $R'=\frac{1}{2}s\bar{I}$, according to Eq. (2.1), is the single-counting rate for an individual detector in the Brown-and-Twiss—type experiment. Obviously, $R'^2\tau_{\text{resp}}$ describes the random coincidence counting rate.

Note that the more $K(0)$ exceeds $R'^2\tau_{\text{resp}}$, the stronger the intensity fluctuates. On the other hand, the equality sign in (2.6) applies only for perfectly amplitude-stabilized light. This explains why thermal light displays a pronounced bunching effect, while light from a single-mode laser does not.

Since we generally can expect that the fluctuations will become statistically independent when τ tends to infinity, we may equate $R'^2\tau_{\text{resp}}$ and $K(\infty)$, which allows us to replace the inequality (2.6) by

$$K(0)>K(\infty) \tag{2.7}$$

for any stationary field displaying intensity fluctuations.

In particular, Eq. (2.3) predicts $K(\tau)$ to have a peak at $\tau=0$. Clearly, this behavior of the coincidence counting rate deserves the name "photon bunching." Hence it becomes evident that the classical wave theory very naturally accounts for the bunching effect. It allows this phenomenon to disappear only for light which is perfectly stabilized in its amplitude, excluding categorically, however, the possibility that the opposite effect, i.e., photon antibunching corresponding to a violation of the inequality (2.3), might occur.

When $K(\tau)$ is a *monotonically* decreasing function of τ, as in the case of thermal light (see Fig. 2), it appears natural to take the normalized quantity

$$\frac{K(0)-K(\infty)}{K(\infty)}=r \tag{2.8}$$

or, since $K(\infty)=R'^2\tau_{\text{resp}}$ under stationary conditions, the relative excess coincidence counting rate (with respect to random coincidences)

$$\frac{K(0)-R'^2\tau_{\text{resp}}}{R'^2\tau_{\text{resp}}}=r \tag{2.9}$$

as a convenient measure of the strength of the bunching effect, the criterion for photon bunching to occur thus being

$$r>0 \; . \tag{2.10}$$

On the other hand, we will speak of photon antibunching when the inverse of (2.10) is true.

In accordance with the majority of workers in the field of photon statistics, we consider the quantity (2.9) as the basic one that characterizes the bunching, or antibunching, properties of any radiation field. This convention is especially attractive for theorists, since it also makes sense in case of single-mode fields that are the preferred objects of theoretical studies concerned with the antibunching phenomenon. Moreover, when based on Eq. (2.9), the criterion (2.10) allows for a natural extension of the bunching/antibunching concept in that it can be applied to an instantaneous state of a *nonstationary* field, as well. To this end, one has to replace time averaging by ensemble averaging. It should be noticed that the averaged values in this case will vary in time. From the experimental point of view it is worth mentioning that a measurement of ensemble averages requires the field to be available in a large number of "copies" produced by a generating mechanism. (An example of such a mechanism is provided by the technique of cw picosecond pulse generation.)

I should like to emphasize that the significant physical quantities that can be determined experimentally with a given degree of accuracy are just ratios of counting rates, rather than their absolute values. Hence the quantity (2.8) is closely connected with observation.

It is noteworthy that both the efficiency and the response time of the detectors cancel out in Eqs. (2.8) and (2.9); this means the quantity r represents a pure property of the radiation field. In the classical wave picture adopted at present, by virtue of Eqs. (2.1) and (2.2), r takes the form

$$r=\frac{\overline{I^2}-\overline{I}^2}{\overline{I}^2}\equiv\frac{\overline{\Delta I^2}}{\overline{I}^2} \; . \tag{2.11}$$

With the observation made above that the antibunching phenomenon is incompatible with the classical wave picture, I am led to consider this effect as a manifestation of a typical corpuscular feature of the radiation field. In fact, I will show in the following section that antibunching is no matter of surprise in a classical particle model.

B. Fluctuating photon numbers

Let us adopt, for the moment, a very simple photon picture, conceiving the photons to be somewhat like point-shaped particles.

It appears natural to assume that the probability of a detector's recording one photon during a time interval $t-\frac{1}{2}\tau_{\rm resp}$ to $t+\frac{1}{2}\tau_{\rm resp}$ is proportional to the number of photons $n(t)$ arriving during that interval at the detector surface. Hence we may write the counting rate, for a given response time $\tau_{\rm resp}$, as

$$R(t)=\beta n(t) \; , \tag{2.12}$$

where the constant β is the detector efficiency.

In general, $n(t)$ will vary in time following the intensity fluctuations of the field.

While the description of the effect the beam splitter exerts on a light beam is a trivial matter in case of light waves, it requires some more attention in the particle picture. We start from the well-known fact that a beam splitter either reflects or transmits, with equal probability, any photon impinging on it. Hence, it splits a "packet" of n incident photons into two packets (corresponding to the reflected and the transmitted beam) which contain k and $n-k$ photons, respectively, where k is one of the numbers $0,1,2,\ldots,n$. Which value of k will be observed in individual circumstances is a matter of chance. We calculate the corresponding probability $w_{k,n-k}$ in a purely classical manner, treating the photons as distinguishable particles interacting independently with the beam splitter.[1] We thus obtain

$$w_{k,n-k}=(\tfrac{1}{2})^n \begin{bmatrix} n \\ k \end{bmatrix} \; . \tag{2.13}$$

[1]Since, *de facto*, photons are neither independent particles nor distinguishable, one might question the validity of formula (2.13). The latter, however, can in fact be substantiated more thoroughly. Starting from a fully quantum-mechanical analysis of light reflection from a plane surface (Brunner, Paul, and Richter, 1965), Paul, Brunner, and Richter (1966) derived the following formula that describes the splitting, due to a partially reflecting mirror, of an incident beam (labeled 0) in an n-photon state, into both a reflected (1) and a transmitted (2) beam

$$|n\rangle_0 \rightarrow \sum_{k=0}^{n} \left[\rho^k(1-\rho)^{n-k}\begin{bmatrix} n \\ k \end{bmatrix}\right]^{1/2} |k\rangle_1 |n-k\rangle_2 \; ,$$

where ρ denotes the mirror reflectivity. [See also the review article by Paul (1966).]

Specifying to a physical situation, where photons are counted, by means of ideal photodetectors, in the reflected and the transmitted beam, respectively, we immediately find from this formula (putting $\rho=\frac{1}{2}$) the expression (2.13) for the probability of detecting k photons in the reflected and $n-k$ photons in the transmitted beam.

It should be emphasized, however, that the quantum-mechanical formula actually accounts for both the particle and the wave aspects of light. In fact, since it ascribes a pure quantum-mechanical state rather than a statistical mixture, to the combined reflected and transmitted field, it predicts specific quantum-mechanical correlations to exist between those two fields which manifest themselves in the capability of the two waves to interfere. In this context, it is interesting to note that the beam splitting experiment in question is of the type of the famous Gedankenexperiment of Einstein, Podolsky, and Rosen (1935), as recently has been pointed out by Paul (1981).

Moreover, it follows from the above quantum-mechanical formula that it would be erroneous to suppose that the numbers of photons in the reflected and the transmitted beam, respectively, have definite, though unknown, values in individual instances. Instead, they must be considered as being intrinsically uncertain. It is the disregard of this specific quantum-mechanical feature that makes the photon picture used in Sec. II.B a classical or naive one.

From Eqs. (2.12) and (2.13) follows the nondelayed coincidence counting rate for the two photocounters in the Brown-and-Twiss—type experiment to be

$$K(t;0)=\beta^2\tau_{\rm resp}\langle\langle \sum_k w_{k,n-k}k(n-k)\rangle\rangle$$
$$=\frac{\beta^2}{4}\tau_{\rm resp}\langle\langle n(t)[n(t)-1]\rangle\rangle , \qquad (2.14)$$

where the double brackets indicate averaging over the ensemble. (When we are dealing with stationary fields, the ensemble average equals the average over a long time.)

On the other hand, the single counting rate for any of the two detectors is given by

$$R'(t)=\beta\langle\langle \sum_k w_{k,n-k}k\rangle\rangle=\frac{\beta}{2}\langle\langle n(t)\rangle\rangle , \qquad (2.15)$$

and the delayed coincidence counting rate for $\tau>\tau_{\rm resp}$ reads

$$K(t;\tau)=\beta^2\tau_{\rm resp}\langle\langle \sum_{k,j} w_{k,n(t)-k}w_{j,n(t+\tau)-j}k(n-j)\rangle\rangle$$
$$=\frac{\beta^2}{4}\tau_{\rm resp}\langle\langle n(t)n(t+\tau)\rangle\rangle . \qquad (2.16)$$

For growing values of τ, the fluctuations in the photon numbers at t and $t+\tau$ will become uncorrelated, i.e., the following relation will hold:

$$K(t;\tau)=R'(t)R'(t+\tau)\tau_{\rm resp} \quad \text{for } \tau\to\infty . \qquad (2.17)$$

From Eqs. (2.14) and (2.15) we find the relative excess coincidence counting rate (2.9) to be

$$r=\frac{\langle\langle n(n-1)\rangle\rangle-\langle\langle n\rangle\rangle^2}{\langle\langle n\rangle\rangle^2} \qquad (2.18)$$

or

$$r=\frac{\Delta n^2-\langle\langle n\rangle\rangle}{\langle\langle n\rangle\rangle^2} , \qquad (2.19)$$

where $\Delta n^2\equiv\langle\langle n^2\rangle\rangle-\langle\langle n\rangle\rangle^2$ is the mean-square deviation of the photon number.

Adopting the criterion (2.10) we infer from Eq. (2.19) that the field has bunching properties when

$$\Delta n^2>\langle\langle n\rangle\rangle , \qquad (2.20)$$

while in the opposite case it displays the antibunching effect.

Since it is well known that a Poisson distribution fulfills the relation (2.20) with the equality sign, we learn that photon antibunching is an attribute of such fields whose photon distribution is narrower than a Poissonian.

Moreover, it becomes obvious from Eq. (2.19) that the antibunching effect will be most pronounced for a beam with a fixed number of photons, $\Delta n^2=0$. (It should be remembered that the photon number, in the present consideration, refers to a volume in the form of a cylinder whose base is given by the detector surface and whose height equals the product of the response time and the velocity of light.)

An experimental realization of a nonfluctuating photon number might be provided by a train of pulses, well separated in space by equal distances cT, each of which contains precisely the same number of photons. The response time of the detector $\tau_{\rm resp}$ should obey the inequality $T>\tau_{\rm resp}\gtrsim\Delta t$, where Δt is the pulse duration. If we measure the time-averaged coincidence counting rates $K(\tau)$ (the averaging procedure extending over a large number of single pulses), the antibunching effect would become manifest in a deficit of $K(0)$ compared to $K(mT)$, where m is any positive integer.

Note that the nonavailability of ideal photocounters (i.e., detectors with 100% detection efficiency) deprives us of the opportunity of directly checking a statement of the type "the photon number in either pulse has the same sharp value." Even when such a physical situation might actually be realized, any realistic measurement will yield a nonzero variance of the number of photons registered. Hence when speaking of fields that contain a fixed number of photons, we either have in mind an idealized situation, in the sense of a Gedankenexperiment, or are assumed to have gained this information from a detailed knowledge of the physical mechanism that generated the field.

It becomes obvious from Eq. (2.18) that the relative excess coincidence counting rate r is independent of the detector efficiency, as in the description of photodetection based on classical electrodynamics. Since a low-efficiency detector might be thought of as being composed of an ideal detector and an absorbing material placed before it, this result leads us to expect that one-photon absorption leaves the characteristic quantity r invariant. In fact, this idea is quickly substantiated more thoroughly by discussing the situation where, not the incident field, but only part of it, separated from it by means of a partly reflecting mirror, is directed to the sensitive area of the photodetector. Actually, the reduction of intensity achieved in this way is physically equivalent to that produced by a normal one-photon absorber. It should be noticed that an essential feature of any attenuation process is that it introduces fluctuations in the photon number when the photon number in the incident field is well defined.

Extension of formula (2.13) valid for a half-silvered mirror to the case of an arbitrary mirror reflectivity ρ gives us the relation (cf. footnote 1)

$$w_{k,n-k}=\rho^k(1-\rho)^{n-k}\binom{n}{k} , \qquad (2.21)$$

from which it follows that

$$\langle\langle k\rangle\rangle=\rho n, \quad \langle\langle k(k-1)\rangle\rangle=\rho^2 n(n-1) , \qquad (2.22)$$

where k is the photon number in the reflected beam and where it has been assumed that the original beam impinging on the mirror contains precisely n photons.

When n itself is a fluctuating quantity, Eqs. (2.22) have to be averaged once more to yield the result

$$\langle\langle k\rangle\rangle=\rho\langle\langle n\rangle\rangle ,$$
$$\langle\langle k(k-1)\rangle\rangle=\rho^2\langle\langle n(n-1)\rangle\rangle . \qquad (2.23)$$

Hence the photon statistical properties of a light beam, as expressed by the quantity r, are identical in the incident and the reflected beam. (The same holds, of course, for the transmitted one.) As a consequence, it suffices to study a part of an original beam, taken from the latter by means of a partly reflecting mirror, in order to determine the photon statistics in the original beam. In this context, it should be noted that the effect of an aperture on a light beam is similar to that of a partly reflecting mirror. Our above result then ensures that photon statistical measurements using detectors with small sensitive areas, in comparison to the beam cross section, are representative for the entire beam, provided, of course, the latter is coherent over its whole cross section.

The physical mechanism underlying the antibunching phenomenon is most easily understood in the case $\Delta n^2=0$. For definiteness, we consider a train of equidistant pulses with the same (sharp) photon number in each pulse. Then the reduction of the nondelayed coincidence counting rate $K(0)$ compared to the coincidence counting rate at a delay time $\tau=mT$ that is a multiple of the difference of the times of arrival for subsequent pulses is readily explained by the following argument: those special events where all the photons contained in one pulse impinging on the beam splitter arrive at one detector, with no photons being left to the second detector, do not contribute to $K(0)$, while they, on the other hand, give nonvanishing contributions to $K(mT)$ in certain cases, which makes the latter quantity greater than the former. Since the events in question become less frequent when the number of photons n per pulse increases, the antibunching effect will decrease in its magnitude for growing n.

Most instructive is the special case $n=1$. Here it is evident that no coincidences can occur for $\tau=0$, while some coincidences certainly will appear for $\tau=mT$.

Comparing bunching and antibunching properties quite generally, we discover a fundamental difference of great physical relevance: photon bunching, as displayed, for example, by thermal radiation, is an intrinsically macroscopic phenomenon. It persists, as a handsome effect, in the limit of arbitrarily high intensities. In fact, since the photons in a (single-mode) thermal radiation field obey Bose-Einstein statistics [see, for example, Glauber (1963b)], Δn^2 equals $\langle\langle n\rangle\rangle^2+\langle\langle n\rangle\rangle$, and hence the relative excess coincidence counting rate (2.19) takes the value $r=1$, irrespective of the mean intensity of the field.

The antibunching effect, on the contrary, bears typical microscopic features, since it disappears for large mean photon numbers $\langle\langle n\rangle\rangle$. Actually, taking into account the fact that antibunching becomes most pronounced for $\Delta n^2=0$, we find from Eq. (2.19) the optimum value for r to be

$$r=-\frac{1}{\langle\langle n\rangle\rangle} \quad (\text{for } \Delta n^2=0) \; . \tag{2.24}$$

Hence, any effort to measure the antibunching effect can be successful at moderate photon numbers, say $\langle\langle n\rangle\rangle \lesssim 100$, only.

The disappearance of the antibunching effect for $\langle\langle n\rangle\rangle\rightarrow\infty$ is felt as satisfactory, on the other hand, from a general point of view. Indeed, it is in accordance with the correspondence principle, which demands that the quantum-mechanical description of the radiation field which gives proper account of the corpuscular nature of light should agree with the classical one in the high-intensity limit.

Certainly, one of the reasons classical electrodynamics precludes the possibility of photon antibunching's occurring is that the description of the detection process, based on the assumption that a photodetector responds to the instantaneous intensity (see Sec. II.A), becomes erroneous at very low intensities, since it conflicts with the energy conservation law. Actually, the detection probability calculated in this way does not vanish for a field that contains less energy than that of a single photon $h\nu$. Specifically in the Brown and Twiss experiment the beam splitter will divide a classical wave packet with energy $h\nu$ into two equal parts, each of which, according to the basic assumption mentioned, might trigger a photocount with a small but nonzero probability. Hence the classical description of the photoelectric detection process allows for coincidences in situations where the energy conservation law strictly forbids them. A theory based on the photon concept, on the contrary, properly accounts for energy conservation and, hence, provides a correct description of photocounting measurements.[2]

However, this is not the whole story. Even when the description of the photoelectric detection process, in the framework of classical electrodynamics, would be modified such as to take proper account of energy conservation, the wave picture would, nevertheless, be in irreconcilable conflict with the photon concept. This is easily seen by asking what will happen when a single photon (in terms of the wave theory, a single pulse with energy $h\nu$) impinges on a beam splitter. Since the wave theory associates the energy with the electric and magnetic field strengths, which involves the concept of the energy's being continuously distributed in space, it predicts that two pulses with energy $\frac{1}{2}h\nu$ stored in each of them will emerge from the beam splitter. In the particle picture, on the contrary, the energy of a photon is an indivisible quantity, which implies that the photon will be either reflected or transmitted as a whole. It then follows from the mere fact that photodetectors respond to the field only when they are supplied with the full energy of one photon $h\nu$ that such instruments intended to measure the reflected and transmitted part, respectively, of the original photon, according to the wave theory will never register a count, whereas the particle theory predicts a single count to be triggered, with a certain probability, at *either* the first *or* the second detector. Hence, convincing

[2]Of course this does not mean that classical wave theory is actually ruled out, since the above-mentioned splitting of wave packets (including those which contain only a single photon) must be invoked, in any case, to explain the interference phenomena.

evidence against the wave picture is already provided by detecting a photon at all in the circumstances under consideration.

Actually, however, such an experiment is extremely difficult to perform—in fact, it has not been realized hitherto—due to the practical impossibility of generating a light beam consisting of single photons well separated from each other so that they will fall on the beam splitter one after the other, but never two, or even more, jointly (i.e., within the response time of the detector). Neither light from a thermal source nor laser radiation meets this requirement. Interestingly, light of the desired type would very distinctly display the antibunching effect, since, obviously, the nondelayed coincidence counting rate, unlike the delayed one, would vanish.

On the other hand, the antibunching effect observed in those specific circumstances would confirm the particle concept of light and contradict the classical wave theory by the mere fact that delayed coincidences are observed at all, since the latter clearly witnesses the triggering of single counts, which contradicts the wave picture, as pointed out above. The typical feature of the antibunching phenomenon as expressed by the inequality $K(0)<K(\infty)$, however, will not be felt as surprising in the ideal case under consideration, because it is a direct consequence of the energy conservation law. Nevertheless, the present discussion underlines how intimately the antibunching phenomenon is connected with the corpuscular aspect of light.

By the way, the above result that the bunching effect strictly disappears, irrespective of the intensity, in case of a Poisson-type photon distribution, leads us to associate with a classical amplitude-stabilized wave a fluctuating photon number governed by Poisson's distribution law. In fact, this suggestion is fully confirmed in the quantum-mechanical formalism, since the photon statistical properties of Glauber states, being the quantum-mechanical analogs of classical waves with fixed phases and amplitudes, are precisely of this kind [cf. Glauber (1963)].

After all, an experimental investigation of the antibunching phenomenon will meet considerable obstacles, the most serious being that none of the light fields generated by present-time sources (including lasers) displays this effect. Hence it will be necessary either to change the photon statistical properties of existing fields—e.g., laser beams—in a definite way, through an appropriate interaction with matter, or to generate light of the desired type directly by controlling the emission process. The first possibility is provided, at least in principle, by nonlinear processes like two-photon absorption or second harmonic generation. In fact, one can expect that such processes tend to smooth fluctuations in the photon number even when the interaction starts from Poisson-distributed photons. The discussion of this problem will constitute one of the main parts of the present article. The second possibility, radically different from the first, utilizes the fact that a single atom being continuously pumped (e.g., by electron collisions or optically via an excitation of higher-lying levels which relax to the upper level of the atomic transition of interest) emits only one photon in every transition. Since the pumping mechanism needs a finite time to excite the atom again after an emission has taken place, the nondelayed coincidence counting rate must vanish, while the delayed one will be different from zero. Indeed, the field generated by a single atom would be a realization of the ideal light beam which plays an essential role in the above-mentioned experiment that would provide the most direct proof of the corpuscular properties of light.

The problem of creating experimental conditions which allow observance of light preferentially from a single atom over a longer period of time has been solved by Kimble, Dagenais, and Mandel (1977) [see also Dagenais and Mandel (1978)] by utilizing an atomic beam technique. Actually, they studied resonance fluorescence rather than spontaneous emission. This first experimental effort to demonstrate the antibunching effect will be described in Sec. VII.B, while the theoretical background for the experiment is discussed in Sec. VI.

Finally, it should be emphasized that the photon statistical analysis presented in this section, however crude the adopted photon picture may appear, is in agreement with a quantum electrodynamical treatment. This will be shown in Sec. II.C.

Before we turn to that, let us observe that, since we used a perfectly classical photon concept, our considerations provide a model for the antibunching effect in the framework of classical mechanics. Specifically, I should like to propose the following realistic device. A "gun" emits, at equidistant instants, packets containing always the same number of balls which, however, differ a little in their propagation direction, the latter being distributed at random over a certain solid angle. The balls impinge on a beam splitter—e.g., a wedge formed by two reflecting plates and positioned such that on the average half of the incident balls are reflected by the first plate, and the other half by the second one. The measurement would consist of counting the number of balls, $n_1^{(\mu)}$ and $n_2^{(\mu)}$, respectively, in the two packets into which the incident packets labelled μ are split ($\mu=1,2,\ldots$). From these findings the averages $\langle\langle n_1^{(\mu)} n_2^{(\mu)}\rangle\rangle$ and $\langle\langle n_1^{(\mu)} n_2^{(\mu+1)}\rangle\rangle$ are to be calculated. The former will be smaller than the latter, and this outcome can be interpreted as a manifestation of the antibunching phenomenon.

C. Quantum-mechanical description

The beam splitter employed by Brown and Twiss (see Fig. 1) may be considered as an elegant experimental solution to the problem of placing two detectors at practically the same position $\mathbf{r}$ within the original beam. Hence the theoretical description of the Brown-and-Twiss—type experiment described in Sec. I essentially reduces to the determination of the probability that the first of two detectors situated at the same position will count a photon during the time interval t to $t+dt$, whereas the second detector will do the same during the time interval $t+\tau$ to $t+\tau+dt$. Using lowest-order perturbation theory, Glauber (1965) has shown this probability, for ideal detectors, to be given by

$$dw^{(2)}=s^2\langle E^{(-)}(\mathbf{r},t)E^{(-)}(\mathbf{r},t+\tau)\times E^{(+)}(\mathbf{r},t+\tau)E^{(+)}(\mathbf{r},t)\rangle(dt)^2 . \quad (2.25)$$

Here, s characterizes the detector sensitivity, and $E^{(+)}$ and $E^{(-)}$ denote the positive and negative frequency part, respectively, of the operator for the electric field strength

$$E^{(+)}(\mathbf{r},t)=\sum_\lambda a_\lambda(\mathbf{r},t)q_\lambda ,$$
$$E^{(-)}(\mathbf{r},t)=\sum_\lambda a^*_\lambda(\mathbf{r},t)q^+_\lambda , \quad (2.26)$$

where the function $a_\lambda(\mathbf{r},t)$ is the (appropriately normalized) classical vector potential for the mode labeled λ, and q^+_λ, q_λ symbolize the familiar photon creation and annihilation operators. The representation (2.26) corresponds to the interaction picture; in particular, the operators q_λ, q^+_λ are time independent in the case of free fields presently under consideration. For simplicity, we have assumed the field to be linearly polarized.

The quantum-mechanical expectation value in Eq. (2.25) represents a special case of the second-order correlation function for the field, which is generally defined as (Glauber, 1963a)

$$G^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2;\mathbf{r}_3,t_3;\mathbf{r}_4,t_4)=\langle E^{(-)}(\mathbf{r}_1,t_1)E^{(-)}(\mathbf{r}_2,t_2)E^{(+)}(\mathbf{r}_3,t_3)E^{(+)}(\mathbf{r}_4,t_4)\rangle . \quad (2.27)$$

Hence the delayed coincidence counting rate in the Brown-and-Twiss—type experiment can be written as

$$K(t;\tau)=\frac{s^2}{4}\tau_{\text{resp}}G^{(2)}(t,t+\tau,t+\tau,t) , \quad (2.28)$$

where the second-order correlation function refers to the field impinging on the beam splitter.

It should be observed that the normal ordering of the operators in Eq. (2.25) ensures the energy's being strictly conserved. Specifically, $dw^{(2)}$ vanishes exactly for a field containing precisely one photon. It is in this respect that the quantum-mechanical formalism proves superior to the classical wave theory.

The counting rate for one detector, on the other hand, is determined by the first-order correlation function which generally reads

$$G^{(1)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)=\langle E^{(-)}(\mathbf{r}_1,t_1)E^{(+)}(\mathbf{r}_2,t_2)\rangle . \quad (2.29)$$

In fact, the probability of a detector's registering a photon in the time interval t to $t+dt$ is [cf. Glauber (1965)]

$$dw^{(1)}=s\langle E^{(-)}(\mathbf{r},t)E^{(+)}(\mathbf{r},t)\rangle dt=sG^{(1)}(\mathbf{r},t;\mathbf{r},t)dt , \quad (2.30)$$

which implies that the single counting rate in the Brown-and-Twiss—type experiment is

$$R'(t)=\frac{s}{2}G^{(1)}(t,t) . \quad (2.31)$$

From Eqs. (2.28) and (2.31) the relative excess coincidence counting rate [cf. Eq. (2.9)] follows:

$$\mathbf{r}(t)=\frac{G^{(2)}(t,t,t,t)-[G^{(1)}(t,t)]^2}{[G^{(1)}(t,t)]^2} , \quad (2.32)$$

where the common argument $\mathbf{r}$ has been omitted for the sake of simplicity. It should be observed that the general expression (2.32) applies also to nonstationary conditions.[3]

As already mentioned in Sec. II.A, the sign of r indicates whether the field has bunching (plus sign) or antibunching (minus sign) properties.

In the framework of quantum electrodynamics, the nonclassical character of the antibunching effect becomes obvious from the fact that the latter does not appear in those fields which possess classical analogs. Formally, fields of such a type are characterized by the property of their density operators ρ to allow for a P representation (Glauber, 1963b)

$$\rho=\int P(\alpha_1,\alpha_2,\ldots)\,|\,\alpha_1,\alpha_2,\ldots\rangle\times\langle\ldots,\alpha_2,\alpha_1\,|\,d^{(2)}\alpha_1 d^{(2)}\alpha_2\ldots \quad (2.33)$$

with a non-negative P function. Here, $|\,\alpha_1,\alpha_2,\ldots\rangle$ denotes a direct product of Glauber states $|\,\alpha_\lambda\rangle_\lambda$ for the modes labeled λ, and the integration runs over the complex α_λ planes for, in principle, all modes of the radiation field ($\lambda=1,2,\ldots$).

The complex number α_λ corresponds to the complex amplitude for the λth mode in the classical description, and the P function can be interpreted as a classical distribution function (provided it is non-negative, which therefore has been assumed above).

By virtue of the well-known formal properties of the Glauber states (Glauber, 1963b),

$$q\,|\,\alpha\rangle=\alpha\,|\,\alpha\rangle ,$$
$$\langle\alpha\,|\,q^+=\langle\alpha\,|\,\alpha^* , \quad (2.34)$$

we find from Eqs. (2.26) the relations

$$E^{(+)}(\mathbf{r},t)\,|\,\alpha_1,\alpha_2,\ldots\rangle=\mathscr{E}(\mathbf{r},t)\,|\,\alpha_1,\alpha_2,\ldots\rangle$$
$$\langle\ldots,\alpha_2,\alpha_1\,|\,E^{(-)}(\mathbf{r},t)=\langle\ldots,\alpha_2,\alpha_1\,|\,\mathscr{E}^*(\mathbf{r},t) , \quad (2.35)$$

[3]From the experimental point of view, an investigation performed under nonstationary conditions would be rather troublesome, since a great number of similar experiments, all starting from the same initial conditions, would then be needed for a measurement of the quantum-mechanical expectation values occurring in Eq. (2.32). Moreover, the random coincidence counting rate $[\frac{1}{2}sG^{(1)}(t,t)]^2\tau_{\text{resp}}$ could be determined only indirectly in this case, via a measurement of the single counting rate $\frac{1}{2}sG^{(1)}(t,t)$.

where

$$\mathscr{E}(\mathbf{r},t)=\sum_{\lambda} a_{\lambda}(\mathbf{r},t)\alpha_{\lambda} \tag{2.36}$$

has the meaning of the positive frequency part of the corresponding classical electric field strength. Hence Eq. (2.32), together with Eq. (2.33), gives us

$$r(t)=\frac{\langle\langle I^2(t)\rangle\rangle-\langle\langle I(t)\rangle\rangle^2}{\langle\langle I(t)\rangle\rangle^2}\ , \tag{2.37}$$

where

$$I(t)\equiv|\mathscr{E}(t)|^2 \tag{2.38}$$

and the symbol $\langle\langle \cdots \rangle\rangle$ denotes averaging with the weighting function P. Obviously, we thus have recovered our previous result (2.11), which relies on a perfectly classical wave concept. Hence we arrive at the conclusion that a field which is "classical," in the sense that it allows for a P representation with a non-negative P function, cannot possess antibunching properties.

It should be noticed, however, that the quantum-mechanical description of those fields differs basically from the classical one by predicting the photon numbers to be fluctuating in any case. Even for a single-mode field in a definite Glauber state, the photon number is not sharp, but instead follows Poisson's distribution law. It is just this property of "classical" fields which reconciles the fact, expressed by Eq. (2.30), that a photodetector on the average responds to the mean intensity with the energy conservation law. Indeed, however small the mean intensity may be in a field characterized by a Glauber state, there exists a nonzero probability that one photon will be present; moreover, even the probability of finding two, three, or more photons does not vanish exactly.

D. Single-mode fields

It is always tempting for a theorist to restrict his analysis to single-mode fields, since this idealization drastically simplifies the theoretical description. Specifically, the relative excess coincidence counting rate (2.32) takes in this case the form

$$r=\frac{\langle q^{+2}q^2\rangle-\langle q^+q\rangle^2}{\langle q^+q\rangle^2}\ . \tag{2.39}$$

This relation is formally identical to the previous formula (2.18) obtained by rather intuitive arguments. There is seemingly a difference in the physical interpretation, since the operator q^+q describes the number of photons contained in the mode volume V, whereas the quantity n in Eq. (2.18) is the photon number with respect to the volume V_{det} defined by the detector parameters (the sensitive area and the response time, see Sec. II.B). However, we have seen in Sec. II.B that a measurement on a "representative" part of the original beam (taken from the latter, in the model calculation, by means of a partly reflecting mirror) suffices to determine the quantity (2.18), where n refers to the original beam. A similar separation is provided, when one is dealing with a single-mode field, by using a detector with $V_{\text{det}}<V$. Indeed, in such a field any correlation, if present, extends over the whole mode volume V, and hence the whole information on the photon statistics can be extracted from a subvolume of V already. Thus we see that there is also a perfect physical equivalence between Eqs. (2.18) and (2.39), provided detectors are used for which $V_{\text{det}}<V$. In particular, Eq. (2.39) reproduces the result (2.24) indicating that antibunching is a $1/\bar{n}$ effect, where $\bar{n}$ is the mean number of photons in the mode volume.

The question arises as to the physical conditions under which a single-mode description will be justified. Usually, one associates a single-mode excitation with a field existing in a suitable cavity. Particularly, it is in this sense that one speaks of single-mode laser operation. However, in the physical processes we shall study later on, we shall generally deal with fields traveling in space. Nevertheless, a single-mode formalism might be employed in this case, too.

From the formal point of view, the situation becomes most transparent when we consider trains of pulses which do not overlap in space—as they are generated in picosecond pulse lasers, for example. A very elegant quantum-mechanical description of a coherent single pulse is due to Titulaer and Glauber (1966), who introduced the concept of nonmonochromatic modes. In the following, I give a short account of their procedure.

Starting from the familiar decomposition of the radiation field into monochromatic modes [see Eqs. (2.26)], they defined nonmonochromatic classical wave packets as superpositions of monochromatic waves $a_\lambda(\mathbf{r},t)$

$$b_l(\mathbf{r},t)=\sum_{\lambda}\gamma_{l\lambda}a_{\lambda}(\mathbf{r},t)\ . \tag{2.40}$$

Here the essential assumption was made that the $\gamma_{l\lambda}$ are elements of a unitary matrix. Note that, anyway, one row of this matrix can be chosen arbitrarily (the only restriction being the normalization condition), i.e., we have the freedom to give at least one of the wave packets a form we like.

Introducing now operators Q_1, Q_1^+ through the definitions

$$Q_l=\sum_{\lambda}\gamma_{l\lambda}^{*}q_{\lambda},\quad Q_l^+=\sum_{\lambda}\gamma_{l\lambda}q_{\lambda}^+\ , \tag{2.41}$$

one may decompose the positive and negative frequency parts of the operator for the electric field strength (2.26) in the form

$$E^{(+)}(\mathbf{r},t)=\sum_{l}b_lQ_l,\quad E^{(-)}(\mathbf{r},t)=\sum_{l}b_l^{*}Q_l^+\ . \tag{2.42}$$

Due to the unitarity of the matrix $(\gamma_{l\lambda})$, the operators Q_l,Q_l^+ have the same formal properties as q_λ,q_λ^+. Hence they can be interpreted as photon annihilation and creation operators as well, the photons now being of nonmonochromatic type, however.

Specializing to the case of only one nonmonochromatic mode being excited, we may disregard the remaining (nonmonochromatic) modes, since they will not contribute to any measurable quantity which, as a general rule, is represented by an expectation value for normally ordered products of photon creation and annihilation operators. In perfect analogy to the familiar case of a monochromatic mode, we find a complete system of eigenstates for the photon number operator Q^+Q by repeatedly applying the photon creation operator to the vacuum state

$$|n\rangle=\frac{1}{\sqrt{n!}}(Q^+)^n|0\rangle \quad (n=0,1,2,\ldots) . \tag{2.43}$$

Thus the formal description of single-mode states of the field is the same for both monochromatic and nonmonochromatic modes. In particular, Eq. (2.39) holds also in case of a wave packet corresponding to a nonmonochromatic mode.

For the theorist, wave packets of rectangular shape (in the direction of wave propagation) are of special convenience, since they correspond to a monochromatic oscillation of the field, which, however, lasts only a finite time, when observed at a fixed position. It is in this way that contact can be made between theoretical studies in which the fields are idealized as monochromatic modes (and most investigations are of this type!) and realistic devices.

It should be emphasized, however, that the single-mode treatment of physical processes relies on the assumption that the single-mode description will apply to the wave(s) involved not only at the beginning, but also in the course of the interaction process. This means that it is presupposed that the shape of the wave packet will not change noticeably during the interaction. This is in no way a trivial assumption. Consider, for example, the parametric three-wave interaction that will be studied in some detail in Sec. IV. When the bandwidth of the incident signal (or idler) pulse exceeds that allowed by the phase matching condition, the outer parts of the Fourier spectrum for the pulse will not be affected by the interaction, which gives rise to a change of the pulse shape. In such a case, the single-mode formalism certainly does not apply.

A specific feature of the single-mode scheme is that any correlation produced in the interaction process will necessarily extend over the whole mode volume—i.e., the formalism is not capable of accounting for an eventual generation of temporal patterns on a time scale that is shorter than the coherence time $\tau_{\rm coh}=l/c$, where l is the length of the mode volume and c the velocity of light. Moreover, it should be kept in mind that the strength of the antibunching effect, by order of magnitude , is given by the inverse of the mean number of photons contained in V, and hence critically depends on the dimensions of V. Consequently, a proper choice of the mode volume, in specific experimental conditions, is actually of physical relevance.

Obviously, this problem lies outside the scope of the single-mode formalism. It requires a multimode treatment of the field, whose basic features will be described in Sec. III.G for the case of two-photon absorption. This analysis will enable us to determine the correlation time for the antibunching effect and to specify the mode volume in terms of physical parameters. In preparation, the basic physical ideas underlying this specification will be discussed in Sec. III.F.

III. MULTIPHOTON ABSORPTION

A. Remarks on one-photon absorption

In what follows, I shall study the effect two-photon, or more generally multiphoton, absorption has on the photon statistics of light, my goal being to demonstrate that multiphoton absorption provides, at least in principle, a means of producing light beams with antibunching properties.

Before that, however, let us briefly consider one-photon absorption. Restricting ourselves to the linear absorption regime—i.e., assuming that the number of atoms that become excited in the course of the absorption process is small compared to the number of atoms in the ground state, we may adopt a simple heat-bath formalism to describe the evolution of the field. When the latter is assumed to be in a single-mode state, the problem thus reduces to that of a damped harmonic oscillator. Then the equation of motion, in the interaction picture, reads [cf. Senitzky (1960, 1961), Lax (1966), or Paul (1969)]

$$\dot{q}^+=-\frac{\kappa}{2}q^+ +F^+ . \tag{3.1}$$

Here κ denotes the damping constant, and F^+ is a fluctuating force (often termed a Langevin force) with the characteristic properties

$$\langle F^+(t_1)F(t_2)\rangle=0 , \tag{3.2}$$

$$\langle F(t_1)F^+(t_2)\rangle=\kappa\delta(t_1-t_2) . \tag{3.3}$$

The expectation value for any product which contains unequal numbers of operators F and F^+, respectively, vanishes. (In particular, $\langle F\rangle$ and $\langle F^+\rangle$ are equal to zero.) Since the fluctuations are of Gaussian character, correlation functions of higher order in F^+,F can be expressed in a well-known manner in terms of products of the first-order correlation functions (3.2) and (3.3) (Wang and Uhlenbeck, 1945). Combined with Eq. (3.2), this implies that the expectation value for any *normally ordered* product of operators F^+,F vanishes.

Equation (3.1) is easily integrated to yield

$$q^+(t)=e^{-(\kappa/2)t}q^+(0)+\int_0^t e^{-(\kappa/2)(t-t')}F^+(t')dt' . \tag{3.4}$$

It becomes obvious from Eq. (3.4), together with what has been said about the Langevin forces, that the latter do not contribute to the expectation value for any *normally ordered* product of photon creation and annihilation operators. Moreover, it follows from Eq. (3.4) that the relative excess coincidence counting rate r [see Eq. (2.39)] remains invariant under one-photon absorption. Note that this result is in perfect agreement with our previous conclusion inferred from a naive photon concept that r is not changed in the process of partial reflection from a mirror.

Hence for a change of the photon statistical properties of light, nonlinear interaction mechanisms will be required. As a first one, we investigate the multiphoton absorption process under this aspect.

B. Basic equations for multiphoton absorption

The first theorists to predict antibunching properties to be generated in two-photon absorption were Chandra and Prakash (1970), who evaluated the density operator for the interacting field up to the lowest order in time-dependent perturbation theory. In the following, I shall outline the master equation approach, allowing, in addition, for more than two photons to be simultaneously absorbed.

For the sake of mathematical simplicity we assume the field to be in a single-mode state[4] corresponding to a finite oscillation at circular frequency ω (and to remain so in the course of interaction), deferring the discussion of the multimode case to Sec. III.G. For the description of k-photon absorption ($k=2,3,\ldots$) we use an effective interaction Hamiltonian of the form (Shen, 1967)

$$H_{\text{int}}=\hbar\sum_{\mu}(\xi_\mu^* a_\mu^+ q^k+\xi_\mu a_\mu q^{+k}) \; . \tag{3.5}$$

Here a_μ^+, a_μ designate the raising and lowering operators which connect the initial and the final states of the μth atom, and ξ_μ is the coupling constant, which, in general, depends on the position of the μth atom. In the case of a running plane wave which we have in mind, this dependence is contained in a phase factor,—i.e., $|\xi_\mu|^2$ is independent of μ.

The advantage of choosing the interaction Hamiltonian in the form (3.5) is, of course, that k-photon absorption associated with excitation of an atom being initially in the ground state and, similarly, k-photon emission accompanied by deexcitation of an excited atom appear as the fundamental processes. This drastically simplifies the formalism compared to that starting from the conventional interaction Hamiltonian, which is linear in q and q^+.

Our primary goal is to eliminate the atomic variables. We start from the equation of motion for the density operator of the entire system $\rho(t)$, which in the interaction picture reads

$$i\hbar\frac{d\rho(t)}{dt}=[H_{\text{int}}(t),\rho(t)] \; . \tag{3.6}$$

From this equation we derive an equation of motion for the density operator for the field, ρ_F, which is defined as

$$\rho_F(t)=\text{Tr}_A\{\rho(t)\} \; , \tag{3.7}$$

where the symbol Tr_A denotes the trace operation over the atomic subsystem. This is achieved in the well-known master equation formalism, which in its simplest mathematical form proceeds as follows [cf. the treatment of nuclear induction by Wangsness and Bloch (1953)]: We assume that at time $t=0$, when the interaction is switched on, the radiation field and the atomic system are decoupled,—i.e., the density operator $\rho(0)$ factorizes in the form

$$\rho(0)=\rho_F(0)\times\rho_A(0) \; , \tag{3.8}$$

where $\rho_A(0)$ designates the density operator for the atomic system at $t=0$. Moreover, we suppose the atoms to be all in their ground states at $t=0$. For this initial condition we solve Eq. (3.6) for a finite time interval τ, using standard perturbation theory up to second order, to find the result

$$\begin{aligned}\rho_F(\tau)-\rho_F(0)=-\tfrac{1}{2}\tau\alpha_k[&q^{+k}(0)q^k(0)\rho_F(0)\\ &-2q^k(0)\rho_F(0)q^{+k}(0)\\ &+\rho_F(0)q^{+k}(0)q^k(0)] \; .\end{aligned} \tag{3.9}$$

In this calculation, strong inhomogeneous line broadening has been assumed to be present—i,e., the atomic frequencies $\omega_A^{(\mu)}=\omega_2^{(\mu)}-\omega_1^{(\mu)}$, where $\hbar\omega_2^{(\mu)}$ and $\hbar\omega_1^{(\mu)}$ denote the energies of the excited and the ground state for the μth atom, respectively, are supposed to be distributed, with a large spread, around a central value ω_{21}, which obeys the resonance condition $\omega_{21}=k\omega$. Strictly speaking, the inhomogeneous linewidth $\Delta\omega_A$ has been assumed to satisfy the condition

$$\Delta\omega_A\tau\gg 2\pi \; . \tag{3.10}$$

The effective coupling constant α_k in Eq. (3.9) is proportional to the spectral density of atoms $\sigma(\omega_A)$, taken at the resonance value $\omega_A=k\omega$,

$$\alpha_k=2\pi\sigma(k\omega)|\xi_\mu|^2 \; . \tag{3.11}$$

(Note that σ refers to all the atoms contained in the mode volume.)

Now, the same procedure is applied to all subsequent time intervals of length τ, as well. This necessitates the nontrivial assumption that decoupling in the form (3.8) takes place at the beginning of *every* time interval. In fact, this assumption, being incompatible with the rigorous solution of Eq. (3.6), constitutes a specific approximation scheme which traces back to Pauli (1928). It should be noted that more sophisticated treatments

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[4]This implies that the polarization of light is uniquely specified. The assumption that two equally polarized photons are simultaneously absorbed becomes problematic for $J=0\rightarrow 0$ transitions, as has been pointed out by Ritze and Bandilla (1980). In the case of circularly polarized light, the presence of both a left- and a right-circularly polarized wave is needed for two-photon absorption to take place, and when the incident light is linearly polarized, a beam polarized in a perpendicular direction will be generated in the course of interaction. The single-mode treatment is correct, however, when at least one of the spins associated with the upper and the lower level of the two-photon transition is different from zero, since then two photons in the same circular polarization state can be jointly absorbed.

based upon projector techniques are free from this unsatisfactory feature. They lead to so-called generalized master equations [cf., for instance, Haake (1973)], which, however, can be solved only approximately. In this way, our final result (3.12) (see below) can be substantiated more thoroughly.

Restricting ourselves to the linear absorption regime—i.e., supposing that the absorption process causes only a negligible population of the upper level, our treatment rests on the assumption that Eq. (3.9) is valid not only for the first time interval but for all subsequent ones, too. Replacing, formally, the difference quotient by the derivative, we finally arrive at the following master equation for the density operator of the field (Shen, 1967):

$$\frac{d\rho_F}{dt}=-\tfrac{1}{2}\alpha_k(q^{+k}q^k\rho_F-2q^k\rho_F q^{+k}+\rho_F q^{+k}q^k)\ , \quad (3.12)$$

where the argument common to all operators is t.

Using the photon number states $|n\rangle$ (Fock states) as a basis in the Hilbert space, one easily recognizes that the evolution of the diagonal elements of the density operator

$$p_n(t)\equiv\langle n\,|\rho_F(t)|\,n\rangle \quad (3.13)$$

is governed by diagonal elements only, but is not affected by off-diagonal elements. Since the quantities $p_n(t)$, being the probabilities of finding n photons in the field at time t (by a suitable measurement) give a full account of the photon statistical properties of the field, it becomes obvious that coherence properties of the field of which off-diagonal elements of the density operator are characteristic, are irrelevant for the change of photon statistics due to multiphoton absorption. Hence we may confine our attention to the diagonal elements (3.13), the equations of motion for which follow from Eq. (3.12) to be

$$\frac{dp_n}{dt}=-\alpha_k\left[\frac{n!}{(n-k)!}p_n-\frac{(n+k)!}{n!}p_{n+k}\right] \quad (n=0,1,2,\ldots)\ . \quad (3.14)$$

Using the familiar transition probability concept, we may readily interpret Eqs. (3.14) as rate equations (Simaan and Loudon, 1975a; Mohr and Paul, 1978). In fact, from standard perturbation theory one finds the probability per unit time for a k-photon absorption process to take place when initially n photons are present to be given by

$$w_{n,n-k}=\alpha_k\,|\langle n-k\,|q^k|\,n\rangle|^2=\alpha_k\frac{n!}{(n-k)!}\ . \quad (3.15)$$

Now, the probability p_n decreases due to a transition from $|n\rangle$ to $|n-k\rangle$, and it increases, on the other hand, on account of a transition from $|n+k\rangle$ to $|n\rangle$. Utilizing Eq. (3.15), we thus immediately arrive at Eqs. (3.14) valid in the linear absorption regime.

C. Rigorous solution

A rigorous solution to the coupled system of equations (3.14) has been found in case of two-photon absorption by introducing a generating function defined as

$$G(s,t)=\sum_{n=0}^{\infty}(1-s)^n p_n(t) \quad (3.16)$$

(Agarwal, 1970; McNeil and Walls, 1974; Tornau and Bach, 1974). The generating function obeys the following partial differential equation:

$$\frac{\partial G}{\partial t}=\alpha_2(2s-s^2)\frac{\partial^2 G}{\partial s^2}\ . \quad (3.17)$$

From $G(s,t)$ one obtains both the probability distribution and the factorial moments by differentiation:

$$p_n(t)=\frac{1}{n!}\left[\frac{\partial}{\partial s}\right]^n G(s,t)\Big|_{s=1}\ , \quad (3.18)$$

$$\langle n^{[l]}(t)\rangle\equiv\langle q^{+l}q^l\rangle=\sum_{n=0}^{\infty}n(n-1)\cdots(n-l+1)p_n(t)=\left[\frac{\partial}{\partial s}\right]^l G(s,t)\Big|_{s=0}\ . \quad (3.19)$$

By virtue of the substitutions

$$x=1-s,\quad T=\alpha_2 t,\quad g(x,T)=G(s,t)\ , \quad (3.20)$$

Eq. (3.17) transforms into

$$\frac{\partial g}{\partial T}=(1-x^2)\frac{\partial^2 g}{\partial x^2}\ . \quad (3.21)$$

This equation can be solved, utilizing the method of separation of variables, to yield

$$g(x,T)=b_0-b_1x+\sum_{n=0}^{\infty}b_{n+2}\frac{(1-x^2)C_n^{(3/2)}(x)}{(n+1)(n+2)}\times\exp[-(n+1)(n+2)T]\ . \quad (3.22)$$

Here, the symbols $C_n^{(3/2)}$ denote ultraspherical polynomials, and the expansion coefficients b_n are determined by the initial values for $g(x,t)$ in the form (Tornau and Bach, 1974)

$$b_0=\tfrac{1}{2}[g(-1,0)+g(1,0)]\ ,$$
$$b_1=\tfrac{1}{2}[g(-1,0)-g(1,0)]\ ,$$
$$b_n=(n-\tfrac{1}{2})\int_{-1}^{+1}[g(x,0)-b_0+b_1x]C_{n-2}^{(3/2)}(x)dx \quad \text{for } n\geq 2\ . \quad (3.23)$$

Making use of Eqs. (3.18) and (3.19), one finally obtains the following expressions for the relevant physical quantities p_m and $\langle n^{[l]}\rangle$ (Tornau and Bach, 1974):

$$p_m(T)=\frac{2^m\Gamma(m-\frac{1}{2})}{m!\Gamma(-\frac{1}{2})}\sum_{n=m}^{\infty}b_nC_{n-m}^{(m-1/2)}(0)\times\exp[-n(n-1)T] \quad (m=0,1,2,\ldots)\ , \quad (3.24)$$

and

$$\langle n^{[l]}(T)\rangle=\frac{2^l\Gamma(l-\frac{1}{2})}{\Gamma(-\frac{1}{2})}\sum_{n=l}^{\infty}b_nC_{n-l}^{(l-1/2)}(1)\exp[-n(n-1)T]\quad(l=1,2,\ldots)\;, \tag{3.25}$$

where

$$C_{n-m}^{(m-1/2)}(0)=\begin{cases}(-1)^{(n-m)/2}\dfrac{\Gamma\left[\dfrac{m+n-1}{2}\right]}{\Gamma(m-\frac{1}{2})\Gamma\left[\dfrac{n-m+2}{2}\right]} & \text{for } n-m \text{ even}\\ 0\quad\text{for } n-m \text{ odd}\;,\end{cases} \tag{3.26}$$

and

$$C_{n-l}^{(l-1/2)}(1)=\begin{bmatrix}n+l-2\\ n-l\end{bmatrix}\;. \tag{3.27}$$

The symbol Γ stands for the gamma function.

Different analytical schemes that allowed derivation of exact solutions of the master equation (3.12) have been developed by Voigt, Bandilla, and Ritze (1980) and Zubairy and Yeh (1980).

However satisfactory it may be from the mathematical point of view to have constructed a rigorous solution, it is still rather troublesome to evaluate formulas like (3.24) and (3.25) in practical cases, the numerical effort drastically increasing with the initial mean photon number $\bar{n}(0)$. In fact, the calculations have been restricted mostly to rather small values of $\bar{n}(0), \bar{n}(0)\leq 20$. As an important result, Tornau and Bach (1974) found photon antibunching to be generated when the absorption process starts from Poisson-distributed photons.

Since, on the other hand, multiphoton absorption produces an observable effect on the field only when its intensity is very high, it is desirable to study the case of large initial mean photon numbers in some detail. This will be done in Sec. III.D by means of a simple approximation scheme which, nevertheless, yields the relevant physical information on the photon statistical properties of the field, as they vary in the course of k-photon absorption ($k=2,3,\ldots$).

D. Approximate solution

Since the physical quantities in which we are immediately interested are the mean photon number $\bar{n}$ and its variance Δn^2, it appears appropriate to pass from Eqs. (3.14) to the equations of motion for the moments of the photon number, defined as

$$\overline{n^l}=\sum_n p_n n^l\quad(l=1,2,\ldots)\;, \tag{3.28}$$

the result being

$$\frac{d\overline{n^l}}{dt}=-\alpha_k\overline{n(n-1)\cdots(n-k+1)[n^l-(n-k)^l]}\;. \tag{3.29}$$

Since the evolution of $\overline{n^l}$ depends on moments of order higher than l (except in the case of one-photon absorption, $k=1$), the hierarchy of equations (3.29) is, of course, no less complicated than the original system (3.14); Eqs. (3.29) allow, however, for a simple approximate solution in case of a photon distribution satisfying the inequalities

$$\bar{n}(t)\gg 1 \tag{3.30}$$

and

$$h(t)\equiv\frac{\Delta n^2(t)}{\bar{n}(t)}\ll\bar{n}(t)\;. \tag{3.31}$$

This means the photon distribution is assumed to exhibit a marked peak at a certain rather large photon number—i.e., it is similar to a Poisson distribution but quite different from the Bose-Einstein distribution characteristic of chaotic light. This treatment especially covers the most important practical case of a (realistic) laser beam impinging on the attenuator.

Because of (3.30) we can approximate the equations of motion for the first and the second moment by

$$\frac{d\bar{n}}{dt}=-k\alpha_k[\overline{n^k}-\tfrac{1}{2}k(k-1)\overline{n^{k-1}}]\;, \tag{3.32}$$

$$\frac{d\overline{n^2}}{dt}=-k\alpha_k[2\overline{n^{k+1}}-k^2\overline{n^k}]\;. \tag{3.33}$$

Moreover , the assumptions (3.30) and (3.31) justify the following approximation:

$$\overline{n^k}=\bar{n}^k+\begin{bmatrix}k\\ 2\end{bmatrix}\bar{n}^{k-2}\Delta n^2\;. \tag{3.34}$$

Retaining on the right-hand sides of Eqs. (3.32) and (3.33) the terms of highest order in $\bar{n}$ only, we arrive at the two equations

$$\frac{d\bar{n}}{dt}=-k\alpha_k\bar{n}^k\;, \tag{3.35}$$

$$\frac{d(\Delta n^2)}{dt}=-2k^2\alpha_k\bar{n}^k(h-\tfrac{1}{2})\;, \tag{3.36}$$

from which the quantitiy h is easily evaluated as a function of $\bar{n}(t)$ (Paul, Mohr, and Brunner, 1976a) in the form

$$h(t)=\left[h(0)-\frac{k}{2k-1}\right]\left[\frac{\bar{n}(t)}{\bar{n}(0)}\right]^{2k-1}+\frac{k}{2k-1}\;. \tag{3.37}$$

Equation (3.37) indicates that for strong attenuation $(\bar{n}(t)/\bar{n}(0)\rightarrow 0)$ h approaches the asymptotic value

$$h_{as}^{(k)}=k/(2k-1) \quad (3.38)$$

(see Fig. 3), this result, for $k=2$, being in excellent agreement with an exact (numerical) calculation by Tornau and Bach (1974).

It is interesting to note that the asymptotic behavior in question is independent[5] of the initial value of h, irrespective of whether $h(0)$ is greater or smaller than h_{as}. Moreover, one learns from Eq. (3.37) that, the higher the number of simultaneously absorbed photons k, the more rapidly the asymptotic value (3.38) is approached, at a given degree of attenuation $\bar{n}(t)/\bar{n}(0)$. This can also be directly seen from Fig. 3.

The result (3.38) implies the relative excess coincidence counting rate (2.39) to tend to the following asymptotic value:

$$r_{as}(t)=\frac{h_{as}-1}{\bar{n}(t)}=-\frac{k-1}{2k-1}\frac{1}{\bar{n}(t)} . \quad (3.39)$$

Obviously, Eq. (3.39) indicates that antibunching occurs as a result of multiphoton absorption $(k=2,3,\ldots)$. The coefficient $(k-1)/(2k-1)$ in Eq. (3.39) takes the value $\frac{1}{3}$ for $k=2$, and it grows only slowly with increasing k, its upper bound being $\frac{1}{2}$. Hence experiments, if feasible at all, will certainly be confined to two-photon absorption. Note that in this case the magnitude of the antibunching effect attains, nevertheless, one-third of the optimum value corresponding to a perfectly sharp photon number [see Eq. (2.24)].

The approximation scheme leading to Eqs. (3.35) and (3.36) has been extended (Mohr and Paul, 1978) to include the third and fourth order moments of the photon number, $\overline{n^3}$ and $\overline{n^4}$, too. Specifically, from $\overline{n^3}$ some information can be gained about the symmetry of the photon distribution curve with respect to the point $n=\bar{n}$. In fact, when the distribution is perfectly symmetric, the value of $\overline{(n-n)^3}$ equals zero. From this reason the quantity

$$\varphi\equiv\overline{(n-\bar{n})^3}(\Delta n^2)^{-3/2} \quad (3.40)$$

has been chosen as a convenient asymmetry parameter, and it could be shown that the modulus of φ decreases in the course of interaction, i.e., for decreasing $\bar{n}$. This means there is a tendency to symmetrize a distribution which is asymmetric in the initial state.

It should be remembered that the results (3.38) and (3.39) have been obtained under the assumption that $\bar{n}$ is large compared to unity even in the final state. When the absorption process reaches its ultimate stage—i,e., when $\bar{n}$ becomes of order k—a new feature appears. In fact, it is readily derived from the basic equations (3.14) in a rigorous manner, that h will undergo a marked change in this case, due to the fact that the field settles down into a steady state [cf. Simaan and Loudon (1975a)]. This must happen, in any case, after a sufficiently long period of time has elapsed, since the absorber ceases to act on the field when fewer than k photons are present. Hence the steady-state photon distribution is of the form $p_n>0$ for $n=0,1,2,\ldots,k-1$ and $p_n=0$ for $n\geq k$.

Since in the hierarchy (3.14) p_n couples to p_{n+k} only, the system (3.14) actually decomposes into k subsystems, the first of which connects all the p_n's for which $n=0\,\mathrm{mod}(k)$, while the second does the same for all the p_n's for which $n=1\,\mathrm{mod}(k)$, and so forth. Evidently, the final photon number corresponding to the first subsystem is zero, the number for the second subsystem is one, etc. Hence the initial probabilities will ultimately "condense" into $p_0(\infty),p_1(\infty),\ldots,p_{k-1}(\infty)$, whose values are given by

$$p_n(\infty)=\sum_{j=0}^{\infty}p_{n+k\cdot j}(0) \quad (n=0,1,2,\ldots,k-1) . \quad (3.41)$$

When the natural assumption is made that the initial photon distribution is smooth, $\bar{n}(0)$ being large compared to unity and Δn^2 being subjected to the *only* condition $\Delta n^2 \gg k^2$, the right-hand side of Eq. (3.41) will, practically, be the same for all n. This gives us (Mohr and Paul, 1978)

$$h(\infty)=(k+1)/6 \quad (k=2,3,\ldots) . \quad (3.42)$$

This result, in particular, indicates that the quantity h, in the course of two-photon absorption, falls from its asymptotic value $\frac{2}{3}$ to its ultimate value $\frac{1}{2}$ when the mean photon number approaches its steady-state value $\bar{n}=\frac{1}{2}$, the relative excess coincidence counting rate thus becoming $r(\infty)=-(2\bar{n})^{-1}=-1$.

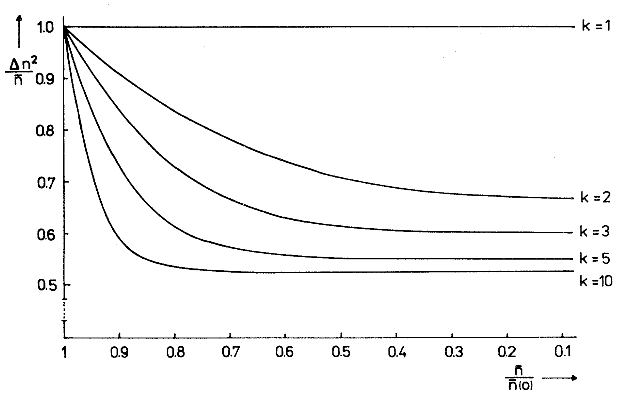

FIG. 3. Change of photon statistics due to k-photon absorption, according to Eq. (3.37). [After Mohr and Paul (1978).]

E. Asymptotic behavior of the field

In the preceding section (III.D) the photon distribution has been shown to display an asymptotic behavior which is characterized by the existence of an asymptotic value

[5]It should be remembered that our proof rests upon the assumptions (3.30) and (3.31). The latter can actually be dropped, however, as will be pointed out in Sec. III.E.

for the quantity $h(t)$ [Eq. (3.31)]. This peculiar feature of the k-photon absorption process has been studied, for $k=2$, more thoroughly by Bandilla and Ritze (1975, 1976a). These authors started from the rigorous solution presented in Sec. III.C. Noticing that the last of the equations (3.23) can be rewritten, by means of partial integration,[6] as

$$b_n = -(n-\tfrac{1}{2})\int_{-1}^{1} g'(x,0)P_{n-1}(x)dx \quad \text{for } n \geq 2 , \tag{3.43}$$

where the prime symbolizes differentiation with respect to x and P_m denotes a Legendre polynomial, they argue as follows: assuming the initial mean photon number to be large compared to unity and, moreover, the initial photon distribution to be smooth, one concludes from the explicit expression for $g'(x,0)$ [cf. Eqs. (3.20) and (3.16)],

$$g'(x,0) = \sum_{n=1}^{\infty} p_n(0) n x^{n-1} , \tag{3.44}$$

that $g'(x,0)$ is rather small compared to unity, in the interval of interest, except in the neighborhood of $x=1$, where it exhibits a marked peak. Hence, for not too great values of $n, n \ll \bar{n}(0)$, it appears justified to replace $P_{n-1}(x)$, in the integrand in Eq. (3.43), by its value at $x=1, P_{n-1}(1)=1$. This gives us, in the case of a smooth initial photon distribution,

$$b_n = -n + \tfrac{1}{2} \quad \text{for } n=2,3,\ldots \ll \bar{n}(0) . \tag{3.45}$$

Moreover, the latter relation holds also for $n=0$ and 1, as follows from Eqs. (3.23), since $g(-1,0)\approx 0$ under the aforementioned condition, and $g(1,0)=1$.

On the other hand, it becomes obvious from Eqs. (3.22), (3.24), and (3.25) that, due to the appearance of the damping factors $\exp[-n(n-1)T]$, the coefficients b_n corresponding to small values of the subscript n become more and more dominating in the course of interaction. Hence, after strong attenuation of the field due to two-photon absorption, the physical quantities of interest are actually determined by coefficients of the form (3.45) only. Since the latter are independent of the specific initial photon distribution, one recognizes that at this stage of the evolution the information on the initial state of the field is completely lost and the field behaves in a unique manner. This statement is true under the above-mentioned assumptions on the initial photon distribution, which are very general. In fact, they imply only the requirement $\Delta n^2(0) \gg 1$ but not the more stringent condition (3.31). Hence the present analysis extends the range of validity, as far as the initial state of the field is concerned, of the results obtained in Sec. III.D, which indicated the existence of an asymptotic behavior of the field. In particular, the incident light field might as well be a chaotic one.[7] In those circumstances and, more generally, in any situation where $\Delta n^2(0) \gg \bar{n}(0)$, multiphoton absorption will, in the first stage, reduce the intensity fluctuations of the field—this effect being fully describable in terms of classical electrodynamics already (Weber, 1971)—until the width of the photon distribution becomes small enough to satisfy the relation $\Delta n^2 = \bar{n}$. It is only in the subsequent stage that the field acquires antibunching properties and that the quantum-mechanical formalism becomes indispensable. This consideration may help us to understand why the fields finally all behave in the same manner, irrespective of their initial states. Of course, the time needed to attain the second stage will be longer, the higher the initial values of $\Delta n^2/\bar{n}$.

Bandilla and Ritze (1976a) calculated also $\Delta n^2(t)/\bar{n}(t)$ by inserting into formulas (3.25), for $l=1$ and 2, the values (3.45) for the coefficients b_n. Their result is in excellent agreement with the approximate solution described in Sec. III.D.

In a recent paper, Voigt, Bandilla, and Ritze (1980) present an exact analytical solution to the master equation for the diagonal elements of the density matrix for the k-photon absorbed field $(k=1,2,3,\ldots)$. Starting from this solution they prove an asymptotic photon distribution, not depending on the initial distribution, to exist when the following conditions are fulfilled: (i) the initial mean photon number $\bar{n}(0)$ is sufficiently large, and (ii) the attenuation is so strong that the square of the resulting mean photon number is small compared to $\bar{n}(0)$.

An approximate expression for the photon distribution which applies to that stage of the evolution where the asymptotic behavior of the field becomes manifest has been given by Paul, Mohr, and Brunner (1976b) [for details see Mohr and Paul (1978)]. They showed that a Gaussian distribution which takes proper account of the relation (3.38),

$$p_n = \left[\frac{2k-1}{k}\right]^{1/2} (2\pi\bar{n})^{-1/2} \exp\left[-\frac{2k-1}{k}\frac{(n-\bar{n})^2}{2\bar{n}}\right] , \tag{3.46}$$

to a good approximation fulfills the basic equations (3.14).

Finally, I should like to mention that the influence of two-photon absorption on the statistical properties of light has been studied also under steady-state conditions. Bandilla and Ritze (1976b) investigated the photon statistics of a single-mode laser field, the laser being equipped with an intracavity two-photon absorber. Chaturvedi, Drummond, and Walls (1977) considered two-photon absorption from a single-mode field existing inside a cavity and being pumped by an external driving field. In both cases the field inside the cavity was shown to exhibit antibunching properties under properly chosen experimental conditions.

[6]In deriving Eq. (3.43) it has been observed that the integral $\int_{-1}^{1} P_{n-1}(x)dx$ vanishes. Hence the term $b_1 x$ in the integral for b_n [see Eq. (3.23)] gives no contribution in Eq. (3.43).

[7]Both this case and that of coherent incident light have been analyzed in more detail by Bandilla and Ritze (1975, 1976a).

F. Specification of the mode volume

The above analysis performed in the single-mode formalism indicated that multiphoton absorption, at least in principle, provides a suitable mechanism of generating light fields with antibunching properties. In order to make contact with realistic experimental situations, we still have to specify the dimensions of the mode volume. For simplicity, we confine ourselves to the case of two-photon absorption. As far as the length of the mode volume (in the direction of light propagation) is concerned, let us visualize the basic physical aspects of the problem in our naive photon picture.

Consider a continuous flux of photons. All photons are supposed to travel with the velocity of light c in the same (x) direction. For definiteness, we assume that the number of photons contained in any volume V (which may be thought of as traveling along with the photons) is governed by Poisson's distribution law. Actually, the size of V can be chosen arbitrarily, since it is a specific feature of the Poisson distribution that the photons contained in any subvolume v of V follow Poissonian statistics if the photons in V do. In fact, when the photons are taken as independent (pointlike) particles, the probability of finding k of them in v when the whole volume is filled with n photons is given by

$$w_k=\left[\frac{v}{V}\right]^k\left[1-\frac{v}{V}\right]^{n-k}\left[\frac{n}{k}\right], \tag{3.47}$$

and with the help of this formula, which closely resembles that describing the effect of a partly reflecting mirror [see Eq. (2.21)], we can readily prove the above statement. As mentioned in Sec. II.B, the situation envisaged corresponds to the presence of a classical wave with fixed amplitude and phase whose quantum-mechanical analog is a Glauber state.

What will happen, when the light beam passes through a two-photon absorber? In the interaction that takes place in this case, the basic event is the simultaneous absorption of two photons by one and the same atom. Of course, it would be unrealistic to take the term "simultaneous" literally; actually, the atom will allow the second photon to arrive with a certain delay, compared to the first one. For the delay time, however, an upper bound, say $\tau_{\rm crit}$, will exist.

Hence the beam will be affected by the absorber in such a way that certain photon pairs which meet the requirement that the distance between the two partners, in x direction, does not exceed the critical length $l_{\rm crit}=c\tau_{\rm crit}$ are removed.[8] Clearly, those elementary processes indirectly give rise also to the disappearance of photon pairs with a distance l greater than $l_{\rm crit}$. However, while all basic events that take place reduce the number of photon pairs with $l \lesssim l_{\rm crit}$, their combined effect on the number of photons pairs with l confined to a particular interval $l_1+l_{\rm crit}\geq l>l_1$ $(>l_{\rm crit})$ will be less. Hence starting from the above-mentioned initial state of the "photon gas" where, in fact, the probability of finding two photons in a distance l is independent of l, the two-photon absorber will change things such that the probability in question, as a function of l, will exhibit a dip at $l=0$ whose half width roughly will equal $l_{\rm crit}$. Due to the uniform propagation of the photons, this spatial pattern will be transformed into a similar temporal structure in a Brown-and-Twiss—type device.

Hence our consideration provides an intuitive understanding of how antibunching comes about in the process of two-photon absorption. Moreover, it indicates that the correlation time for the antibunching effect is intimately connected with a memory mechanism in the atoms that enables an individual atom to jointly absorb two photons even when they arrive at different instants. Of course, the determination of the memory time needs a special investigation—which will be performed in Sec. III.G. Evidently, in order to take proper account of the correlation time in the single-mode formalism (in which any correlation, if present, necessarily extends over the whole mode volume), we have to identify the length of the mode volume with the critical length $l_{\rm crit}$.

Now, the question arises as to the cross-sectional area of the mode volume. Before trying to give an answer, I will make some general remarks. To begin with, we notice that the interaction of a single atom with the field is a local one. (Strictly speaking, the interaction extends over the atomic dimensions, which, however, are negligibly small in comparison to the wavelength at optical frequencies.) Hence two photons that become jointly absorbed in an elementary process are both required to arrive at practically the same point in space where the absorbing atom is situated. Therefore, in case a monochromatic plane wave with fixed amplitude and phase is incident (quantum mechanically, such a field being represented by a Glauber state), one might suspect that short-range correlations of antibunching type are produced in the course of interaction with the atoms in a lateral direction. It is a well-known consequence of the wavelike nature of radiation, however, that transverse field correlations quite generally tend to quickly spread over larger and larger areas as the wave propagates farther.

This effect plays an important role in stellar interferometry [see, for instance, Mandel and Wolf (1965)]. It gives rise to the fact that the stellar light, in a narrow frequency band, when arriving on Earth exhibits spatial correlations which extend on the Earth's surface over tens or hundreds of meters, while the original correlation produced on the surface of the star by the incoherent emission process has an extremely short range. (In the calculation, the spatial dependence of the first-order correlation function on the star's surface is assumed to be given by a delta function.) Actually, the transverse coherence length of the radiation, $l_{\rm trans}$, on Earth sensitively depends on the angular diameter α of the star.

[8]Besides, the two photons involved in an elementary absorption process are required to "hit" the same atom. (See the end of this section.)

The technique of stellar interferometry, introduced by Michelson and later on ingeniously improved by Brown and Twiss, utilizes just this relationship, which approximately reads

$$l_{\text{trans}} \approx \frac{\lambda R}{2\rho} = \frac{\lambda}{\alpha} , \tag{3.48}$$

where λ is the wavelength, ρ the radius of the star (idealized as a uniformly radiating disk), and R the distance from the star to the observer.

Turning now to the problem of how to specify the cross-sectional area of the mode volume, we might use the following argument. Clearly, in the single-mode treatment any elementary absorption process affects the electric field at every point within the mode volume in the same manner. This idealization finds a certain justification, as far as the transverse dimensions of the mode volume are concerned, when we determine the lateral dimensions of the mode volume by the following requirement: the area over which a distortion of the wave due to an individual absorption process taking place near the entrance surface of the absorber spreads on the exit surface be equal to the cross section of the mode volume. (In what follows, we assume the absorber length L to exceed that of the mode volume.) Hence an estimate of the magnitude of the spreading effect in question is needed.

We find it in a simple way by utilizing the Huygens-Fresnel priniciple. According to the latter, the field amplitude at a point P_1 originates from the combined action of all the elementary (spherical) waves which started in forward direction, at properly retarded times, respectively, from every point of a given wave front, which is a plane F perpendicular to the direction of wave propagation in the case of an infinitely extended, monochromatic plane wave. However, most of the elementary waves mentioned will interfere destructively at P_1. To see this in more detail, it is useful to construct Fresnel zones in F that are centered at the projection P_1' of P_1 on F (see Fig. 4). Then it can be shown (Grimsehl, 1962; see also Born and Wolf, 1964) that, for an ideal plane wave, the contributions from the outer half of the nth zone, the total $(n+1)$th zone and the inner half of the $(n+2)$th zone $(n=1,2,\dots)$ add up to zero. Hence the amplitude at P_1 can be supposed to be produced, through constructive interference, by only those elementary waves whose centers cover the inner half A_1 of the first Fresnel zone.

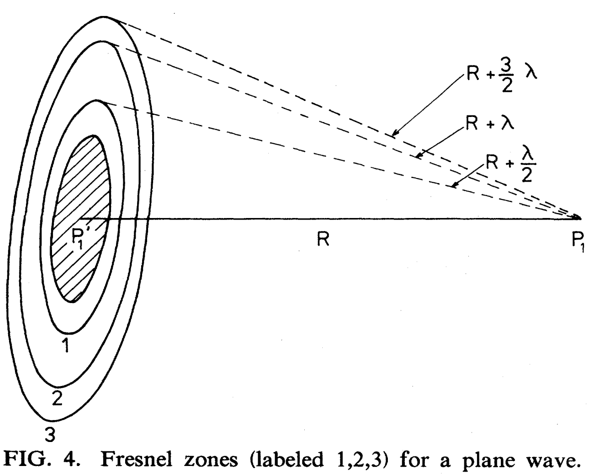

FIG. 4. Fresnel zones (labeled 1,2,3) for a plane wave. The amplitude at P_1 can be supposed to be due to the contributions from the inner half of the first Fresnel zone (hatched area) only.

Obviously, this statement remains valid when the wave amplitudes in F, which determine the amplitudes of the respective elmentary waves starting from F, exhibit a localized distortion (produced, for example, by interaction with matter), provided the area of distortion falls in A_1. What can be concluded from this is the following: suppose that the amplitude is distorted in the neighborhood of P_1' and consider a second point P_2 which has the same distance from F as P_1. Then, the amplitudes at P_1 and P_2 will practically be equal when the area of distortion is contained in the inner half A_2 of the first Fresnel zone corresponding to P_2, as well (see Fig. 5). Otherwise, the two amplitudes will be different. Hence, assuming the area of the distortion in F to be small compared to A_1, we find the radius ρ of the area over which the distortion spreads, due to wave propagation from P_1' to P_1, to be approximately given by the distance between P_1 and P_2 under the condition that the center of A_2 falls on the boundary of A_1. This means that ρ equals the radius of A_1. Utilizing the well-known formula for the radius of the first Fresnel zone, $r_1 = \sqrt{R\lambda}$, where R is the distance between P_1' and P_1, we obtain

$$\rho = \frac{1}{\sqrt{2}} r_1 \equiv \frac{1}{\sqrt{2}} \sqrt{R\lambda} . \tag{3.49}$$

Thus, adopting the above reasoning concerning the transverse dimensions of the mode volume, we find the latter to be given by

$$d_{\text{mod}} = 2\rho = (2L\lambda)^{1/2} . \tag{3.50}$$

The dimension defined by Eq. (3.50) is a manifestly macroscopic one; for example, d_{mod} takes the value 10^{-2} cm for $\lambda = 5\times10^{-5}$ cm and $L=1$ cm. It should be noticed that, in case one employs a light beam whose diameter d at the entrance surface of the interaction volume is much smaller than d_{mod}, this beam will become drastically widened, due to diffraction, while it travels through the

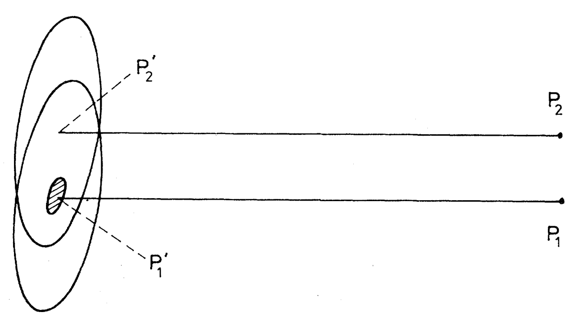

FIG. 5. The inner halves of the first Fresnel zones corresponding to the points P_1 and P_2, respectively. The field is assumed to be distorted in the hatched area.

interaction volume. In fact, even for $d = d_{mod}$, the beam diameter at the exit surface of the interaction volume will be enhanced by a factor of about 2 compared to its value at the entrance surface d.

In the case $d \gg d_{mod}$ it must be ensured experimentally that the detectors register only photons that for zero delay time belong to a single-mode volume. This is achieved by focusing only part of the beam cross section at the exit surface of the interaction volume, namely, an area whose linear dimensions equal d_{mod}, on the sensitive surfaces of the detectors.

The result (3.50) has the following implication for an optimum choice of the experimental conditions. In order to enhance the effectiveness of the two-photon absorption mechanism in producing antibunching properties, it is more favorable to have a higher density of the atoms rather than a greater length of the interaction volume, since in the latter case one must put up with a larger mode volume, which leads to a greater photon number per mode volume at given intensity.

Finally, I should like to emphasize that the choice of the transverse dimension of the mode volume as proposed above is in no way specific to the interaction mechanism; instead, the result (3.50) applies equally well to processes different from multiphoton absorption when a single-mode formalism is used to describe them.

The length of the mode volume, in contrast, is clearly determined by the specific physical features of the interaction process. Hence, this problem must be treated separately. In the following, we shall tackle it for the case of two-photon absorption.

G. Two-photon absorption of a nonmonochromatic field

In order to determine the correlation time for the antibunching phenomenon, we must dispense with the single-mode formalism. In the following, we will use the electric field strength for the description of the field, without decomposing it into contributions from different modes. We consider a plane-wave—type light beam of finite bandwidth which is coherent over its cross section and travels in the x direction. The absorption cell is assumed to be homogeneously filled with atoms capable of two-photon absorption. (The generalization to k-photon absorption is straightforward.) Then the problem may be idealized as one dimensional (with respect to space).

It is well known that the operator for the electric field strength can be written as

$$E(x,t) = E^{(-)}(x,t) + E^{(+)}(x,t) , \qquad (3.51)$$

where the terms on the right-hand side are the negative and positive frequency parts, respectively. For the following it will be convenient to separate from $E^{(\pm)}$ the rapid oscillation in space—i.e., to put

$$E^{(-)}(x,t) = e^{-ik_0x}\hat{E}^{(-)}(x,t) ,$$
$$E^{(+)}(x,t) = e^{ik_0x}\hat{E}^{(+)}(x,t) , \qquad (3.52)$$

where k_0 is the wave number corresponding to the center frequency ω_0. The spatial dependence of $E^{(\pm)}$ is then due to the interaction only.

Generalization of Eq. (3.5) gives us the interaction Hamiltonian in the form

$$H_{\text{int}} = \hbar\xi \sum_{\mu} \hat{E}^{(-)^2}(x_\mu) a_\mu + \text{H.c.} \qquad (3.53)$$

Allowing for both homogeneous and inhomogeneous line broadening, we ascribe to the atoms a (common) homogeneous linewidth 2γ and different resonance frequencies Ω_μ. Since arbitrary phase factors can be included in the lowering and raising operators a_μ and a_μ^+, we were free to take the coupling constant ξ as a real quantity independent of μ.

From the formal point of view, it is desirable to consider the atomic operators—like the field operators—as *continuous* functions in space. Doing so, we rewrite Eq. (3.53) as

$$H_{\text{int}} = \hbar\xi \sum_j p_j \int_0^L dx\, \hat{E}^{(-)^2}(x) a_j(x) + \text{ H.c.} \qquad (3.54)$$

Here, the subscript j has been used to label groups of atoms which differ by their resonance frequencies Ω_j. The factor p_j denotes the number of atoms per unit length in x direction in the jth group, and L is the length of the absorption cell.

The atomic operators $a_j(x), a_j^+(x)$ satisfy the anticommutation relation

$$a_j(x')a_j^+(x) + a_j^+(x)a_j(x') = \frac{1}{p_j}\delta(x-x') . \qquad (3.55)$$

Their commutator, on the other hand, reads

$$a_j^+(x)a_j(x') - a_j(x')a_j^+(x) = \frac{\sigma_j(x)}{p_j}\delta(x-x'), \qquad (3.56)$$

where the operator $\sigma_j(x)$ represents the inversion associated with one atom. In what follows, we shall approximate $\sigma_j(x)$ by -1, thus disregarding saturation effects.

Taking into account only those modes of the field which propagate strictly in the (positive) x direction, we find the commutator for the field operators $E^{(+)}$ and $E^{(-)}$ to be

$$[\hat{E}^{(+)}(x), \hat{E}^{(-)}(x')] = 2\pi\hbar\omega_0\delta(x-x') - 2\pi i\hbar c\delta'(x-x') , \qquad (3.57)$$

where δ' is the derivative of Dirac's delta function, and c is the velocity of light.

From Eqs. (3.54), (3.56), and (3.57) we obtain the equations of motion in the Heisenberg picture. After separation of the high-frequency time dependence of the operators and inclusion of damping terms characteristic of homogeneous line broadening together with associated Langevin forces $\tilde{f}_j^+(x,t)$ [see, for instance, Lax (1966)], they take the following form:

$$\dot{\tilde{a}}_j^+(x,t) = i(\Omega_j - 2\omega_0)\tilde{a}_j^+(x,t) - \gamma\tilde{a}_j^+(x,t) + i\xi\tilde{E}^{(-)^2}(x,t) + \tilde{f}_j^+(x,t) , \qquad (3.58)$$

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\widetilde{E}^{(-)}(x,t)$$
$$=4\pi i\hbar\omega_0\xi\sum_j p_j\widetilde{a}_j^{+}(x,t)\widetilde{E}^{(+)}(x,t)\ , \quad (3.59)$$

where

$$\widetilde{E}^{(-)}(x,t)=e^{-i\omega_0 t}\hat{E}^{(-)}(x,t)$$
$$=e^{i(k_0x-\omega_0 t)}E^{(-)}(x,t)\ , \quad (3.60)$$
$$\widetilde{a}_j(x,t)=e^{2i\omega_0 t}a_j(x,t)\ . \quad (3.61)$$

The symbol γ stands for the damping constant, which is the inverse of the transverse relaxation time (or dephasing time), and it has been assumed that the atomic line center coincides with twice the midfrequency of the field. In deriving Eq. (3.59) only the dominant first term on the right-hand side of Eq. (3.57) has been taken into account.

Integrating Eq. (3.58) and substituting the result into Eq. (3.59), we obtain

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\widetilde{E}^{(-)}(x,t)$$
$$=-4\pi\hbar\omega_0\xi^2\sum_j p_j$$
$$\times\int_{-\infty}^{t}e^{[i(\Omega_j-2\omega_0)-\gamma](t-t')}\widetilde{E}^{(-)2}(x,t')dt'\cdot\widetilde{E}^{(+)}(x,t)$$
$$+\widetilde{F}^{+}(x,t)\widetilde{E}^{(+)}(x,t)\ , \quad (3.62)$$

where

$$\widetilde{F}^{+}(x,t)=4\pi i\hbar\omega_0\xi\sum_j p_j\int_{-\infty}^{t}e^{[i(\Omega_j-2\omega_0)-\gamma](t-t')}$$
$$\times\widetilde{f}_j^{+}(x,t')dt'\ . \quad (3.63)$$

Here we have assumed that the time which elapsed after the beginning of the interaction is large compared to γ^{-1}. Hence the initial values of the operators $\widetilde{a}_j,\widetilde{a}_j^{+},\widetilde{f}_j,\widetilde{f}_j^{+}$ are damped out at time t, and the lower limit of integration can be replaced by $-\infty$, as has been done above.

Supposing the inhomogeneous line to be of Lorentzian shape (with linewidth

$$\sum_j p_j\cdots\to p_{\rm tot}\Gamma\pi^{-1}\int d\Omega[(\Omega-2\omega_0)^2+\Gamma^2]^{-1}\cdots$$

where $p_{\rm tot}$ is the total number of atoms per unit length. The integration over Ω is then easily performed to yield (Mohr and Paul, 1979)

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\widetilde{E}^{(-)}(x,t)$$
$$=-4\pi\hbar\omega_0\xi^2 p_{\rm tot}$$
$$\times\int_{-\infty}^{t}e^{-(\Gamma+\gamma)(t-t')}\widetilde{E}^{(-)2}(x,t')dt'\cdot\widetilde{E}^{(+)}(x,t)$$
$$+\widetilde{F}^{+}(x,t)\widetilde{E}^{(+)}(x,t)\ . \quad (3.64)$$

This equation displays a typical memory effect. It indicates that what happens with the field at a given point x at time t is affected by the electric field strength which existed, at the same point, at previous times t' satisfying the inequality $t-t'\lesssim(\Gamma+\gamma)^{-1}$. The underlying physical mechanism is that the field induces coherent atomic oscillations at the two-photon resonance frequency which add up to a coherent macroscopic oscillation. Once excited, the latter persists for some time, and in this way the field interacting with the oscillation at a given time is influenced by its own behavior in the past.

Clearly, the effective memory time

$$\tau_{\rm mem}=\frac{1}{\Gamma+\gamma} \quad (3.65)$$

has to be identified with the critical time $\tau_{\rm crit}$ we introduced in Sec. III.F—in our naive photon picture—as the upper bound for the delay time between two photons that must not be exceeded by them in order for them to have a chance of getting jointly absorbed by one atom. Hence the desired specification of the length of the mode volume, l, is provided by the relation

$$l=c\tau_{\rm mem}=\frac{c}{\Gamma+\gamma}\ . \quad (3.66)$$

Strictly speaking, this result applies to the case that the linewidth of the radiation $\Delta\omega$ is shorter than $\Gamma+\gamma$. Hence, in those circumstances it is not the coherence time $\tau_{\rm coh}=(2\Delta\omega)^{-1}$, but the shorter atomic memory time which determines what has to be taken as the mode volume. In the opposite case $\Delta\omega\gtrsim\Gamma+\gamma$, the coherent interaction between the atoms and the field will be confined to time intervals whose duration is given by $\tau_{\rm coh}$, due to the random changes the phase of the field undergoes when a time of the order of the coherence time has elapsed.

It seems worth noticing that subjecting Eq. (3.64) as a whole to the temporal averaging procedure

$$\int_{-\infty}^{t}dt'(\Gamma+\gamma)\exp[-(\Gamma+\gamma)(t-t')]\cdots$$

leads to an equation of motion that corresponds to a single-mode treatment of the field (Mohr and Paul, 1979). In contrast to the conventional *ab initio* single-mode formalism, however, this procedure yields a physical definition of the mode volume, as given by Eq. (3.66).

It can also be shown more directly that the correlation time for the antibunching effect equals the memory time (3.65). To this end, we make use of the fact that an equation of motion can be derived from Eq. (3.64) for any correlation function of interest. Fortunately, the fluctuating forces $\widetilde{F},\widetilde{F}^{+}$ give only vanishing contributions in this case. Indeed, it has been shown by Mohr (1981a) that this is a general rule valid for the evolution of any *normally ordered* product of operators $\widetilde{E}^{(-)},\widetilde{E}^{(+)}$.

Specifically, from Eq. (3.64) we obtain

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\langle\widetilde{E}^{(-)}(t)\widetilde{E}^{(+)}(t)\rangle=-\alpha\int_{-\infty}^{t}e^{-(\Gamma+\gamma)(t-t')}\langle\widetilde{E}^{(-)2}(t')\widetilde{E}^{(+)2}(t)\rangle dt'+\text{c.c.}\ , \quad (3.67)$$

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\langle \tilde{E}^{(-)}(t)\tilde{E}^{(-)}(t+\tau)\tilde{E}^{(+)}(t+\tau)\tilde{E}^{(+)}(t)\rangle$$
$$=-\alpha\int_{-\infty}^{t} e^{-(\Gamma+\gamma)(t-t')}\langle \tilde{E}^{(-)2}(t')\tilde{E}^{(+)}(t)\tilde{E}^{(-)}(t+\tau)\tilde{E}^{(+)}(t+\tau)\tilde{E}^{(+)}(t)\rangle dt'$$
$$-\alpha\int_{-\infty}^{t+\tau} e^{-(\Gamma+\gamma)(t+\tau-t')}\langle \tilde{E}^{(-)}(t)\tilde{E}^{(-)2}(t')\tilde{E}^{(+)2}(t+\tau)\tilde{E}^{(+)}(t)\rangle dt'+\text{c.c.}\ , \tag{3.68}$$

or, establishing normal order[9] in the first term on the right-hand side of Eq. (3.68),

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]\langle \tilde{E}^{(-)}(t)\tilde{E}^{(-)}(t+\tau)\tilde{E}^{(+)}(t+\tau)\tilde{E}^{(+)}(t)\rangle$$
$$=-\alpha[\tilde{E}^{(+)}(t),\tilde{E}^{(-)}(t+\tau)]\int_{-\infty}^{t} e^{-(\Gamma+\gamma)(t-t')}\langle \tilde{E}^{(-)2}(t')\tilde{E}^{(+)}(t+\tau)\tilde{E}^{(+)}(t)\rangle dt'$$
$$-\alpha\left[\int_{-\infty}^{t} e^{-(\Gamma+\gamma)(t-t')}\langle \tilde{E}^{(-)2}(t')\tilde{E}^{(-)}(t+\tau)\tilde{E}^{(+)}(t)\tilde{E}^{(+)}(t+\tau)\tilde{E}^{(+)}(t)\rangle dt'\right.$$
$$\left.+\int_{-\infty}^{t+\tau} e^{-(\Gamma+\gamma)(t+\tau-t')}\langle \tilde{E}^{(-)}(t)\tilde{E}^{(-)2}(t')\tilde{E}^{(+)2}(t+\tau)\tilde{E}^{(+)}(t)\rangle dt'+\text{c.c.}\right]. \tag{3.69}$$

Here, α stands for the positive constant

$$\alpha=4\pi\hbar\omega_0\xi^2 p_{\text{tot}}\ , \tag{3.70}$$

and the common argument x has been omitted in order to simplify the notation.

By virtue of Eqs. (3.67) and (3.69), the relative excess coincidence counting rate, which in case of delayed coincidences reads

$$r(x;t,t+\tau)\equiv\frac{\langle \tilde{E}^{(-)}(x,t)\tilde{E}^{(-)}(x,t+\tau)\tilde{E}^{(+)}(x,t+\tau)\tilde{E}^{(+)}(x,t)\rangle}{\langle \tilde{E}^{(-)}(x,t)\tilde{E}^{(+)}(x,t)\rangle\langle \tilde{E}^{(-)}(x,t+\tau)\tilde{E}^{(+)}(x,t+\tau)\rangle}-1\ , \tag{3.71}$$

is readily found to evolve according to

$$\left[\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right]r(x;t,t+\tau)=-\alpha\frac{[\tilde{E}^{(+)}(x,t),\tilde{E}^{(-)}(x,t+\tau)]}{\langle \tilde{E}^{(-)}(x,t)\tilde{E}^{(+)}(x,t)\rangle\langle \tilde{E}^{(-)}(x,t+\tau)\tilde{E}^{(+)}(x,t+\tau)\rangle}$$
$$\times\int_{-\infty}^{t} e^{-(\Gamma+\gamma)(t-t')}\langle \tilde{E}^{(-)2}(x,t')\tilde{E}^{(+)}(x,t+\tau)\tilde{E}^{(+)}(x,t)\rangle dt'+A \tag{3.72}$$

[cf. Mohr (1981a)], where the term abbreviated by A has the important property that it vanishes precisely in case the field is a wave packet with a coherence length greater than τ, represented by a Glauber state.

Hence, it becomes evident that the generation of antibunching properties which formally correspond to a negative value of r is intimately connected with the appearance of the commutator in Eq. (3.72). The latter has its root in the occurrence, in Eq. (3.68), of an expectation value for a *not* normally ordered product of operators $\tilde{E}^{(-)},\tilde{E}^{(+)}$. This result, in fact, does not come as a surprise, but underlines once again the intrinsically quantum-mechanical nature of the antibunching phenomenon.

Since the τ dependence of the commutator in Eq. (3.72) determines the correlation time for the antibunching effect produced in two-photon absorption [when the field is initially in a Glauber state, the quantity r will not change, according to Eq. (3.72), for such values of τ for which the commutator has fallen to zero], the question as to what explicit expression should be inserted for the commutator is of physical relevance.

To take simply the free-field value for the commutator, which, in analogy to Eq. (3.57), displays a δ-function—type singularity at $\tau=0$, appears to be rather unsatisfactory, since this would imply ascribing to the antibunching effect an infinitely short correlation time. In practice, this would mean the correlation time actually observed would be determined by the response time τ_{resp} of the detectors used. In fact, due to the finite value of τ_{resp}, the delay time is specified with an uncertainty of the order of τ_{resp} only. Accordingly, the singularity of the field commutator is "smeared out" over a time interval of that length.

Physically, one will expect, however the correlation time in question to be determined by parameters that are associated with the interaction mechanism. There are two time scales inherent in the physical problem under consideration,

(a) the coherence time of the incident field τ_{coh}, and
(b) the memory time connected with the elementary two-photon absorption process; and it has to be clarified which of them defines the correlation time.

[9]We tacitly assume that the commutator $[\tilde{E}^{(+)}(t),\tilde{E}^{(-)}(t+\tau)]$ is a c number also in case of the interacting field under consideration.

Formally, a finite time constant comes into play when the frequency integration in the evaluation of the (free-field) commutator

$$[\widetilde{E}^{(+)}(x,t),\widetilde{E}^{(-)}(x,t')]=\frac{\hbar}{c}\int_0^\infty \omega e^{i(\omega-\omega_0)(t'-t)}d\omega \tag{3.73}$$

is restricted to a finite interval centered at ω_0—i.e., by introducing a cutoff function of the type $\exp(-T\,|\,\omega-\omega_0\,|\,)$. In this way the commutator (3.73) becomes a well-behaved function exhibiting a peak at $\tau=0$ of half width $2T$. Note that the peak height varies as T^{-1}. According to Eq. (3.72), this implies also that the magnitude of the antibunching effect produced in the interaction, for a given absorber and a fixed intensity of the incident (coherent) light, depends sensitively on the cutoff parameter T.

The essential question is how to choose the value of T or, equivalently, of the effective bandwidth $\Delta\omega\approx T^{-1}$ that defines the frequency range for the modes being admitted to contribute to the commutator (3.73). In my opinion, this problem is resolved by the following argument: consider an incident light wave with a coherence time much greater than the memory time $\tau_{\rm mem}$. Due to the presence of the memory mechanism, the absorbing material, at given x and t, is affected by the electric field strength existing at x in the time interval $t-\tau_{\rm mem}$ to t. Hence, the field appears to the medium as if it were a pulse whose duration is of order $\tau_{\rm mem}$. Under the above assumption $\tau_{\rm coh}\gg\tau_{\rm mem}$, this means that what happens in the absorber in an interval that is short compared to the coherence time cannot depend on the latter. This reasoning rules out the possibility of putting T equal to $\tau_{\rm coh}$. Thus we are led to identify T with the memory time. Since T defines, via Eq. (3.72), the correlation time for the antibunching effect, this means that this correlation time is given by the memory time. This result being precisely what has been suggested in Sec. III.F by rather intuitive arguments, I have substantiated the latter discussion more thoroughly; at least, I hope to have done so.

Needless to say, in order to actually measure the correlation time in question, detectors are needed with a response time shorter than this time. Otherwise, the effect under observation will be more or less wiped out.

Finally, I should like to mention that the analysis performed in this section has been extended by Mohr (1981a, b) to the k-photon case ($k=3,4,\ldots$). Moreover, saturation effects have been discussed in those papers. As a result, the latter have been found to tend to diminish the magnitude of the antibunching effect.

IV. PARAMETRIC THREE-WAVE INTERACTION

A. The degenerate process

It has been shown by several authors that optical parametric three-wave processes in which the medium plays only the role of a "catalyst," are also suited to endow light beams with antibunching properties[10] (Stoler, 1974; Kielich, Kozierowski, and Tanaś, 1977; Kozierowski and Tanaś, 1977; Mišta and Peřina, 1978; Mostowski and Rzążewski, 1978; Trung and Schütte, 1978; Drummond, McNeil, and Walls, 1979; Neumann and Haug, 1979). The main features of this type of interaction's giving rise to changes in the photon statistics become evident from the pioneering work of Stoler (1974), who studied the degenerate parametric three-wave process from the photon statistical point of view. In the following, I present a simple theoretical analysis of this problem.

We are concerned with the following physical situation. A weak optical field at the fundamental frequency ω is passed, together with an intense harmonic at frequency 2ω, through a suitable nonlinear crystal. We assume the phase-matching condition to be fulfilled and the relative phase $\varphi_{2\omega}-2\varphi_\omega$ to be adjusted such that the fundamental wave becomes attenuated, whereas the harmonic undergoes amplification. Hence the process might be termed "stimulated second harmonic generation." Since the basic event is the "fusion" of two photons from the fundamental wave into one second harmonic photon, this process closely resembles, as far as the fundamental wave is concerned, two-photon absorption, and hence one will expect that the changes in the photon statistics of the fundamental wave are of the antibunching type, too.

To make the formal treatment as simple as possible, we idealize the fundamental wave by a single-mode state of the (quantized) radiation field, deferring the problem of specifying the longitudinal dimension of the mode volume to Sec. IV.C. The harmonic, one the other hand, is assumed to be so intense that a classical description will apply, and, moreover, the relative increase of its intensity due to the interaction process can be neglected. This means we treat the harmonic as a given classical plane-wave—type monochromatic wave.

The basic equations of motion for the degenerate parametric process may be written, in the interaction picture, as [cf. Louisell, Yariv, and Siegman (1961), Mollow and Glauber (1967), Brunner and Paul (1977)][11]

$$\dot{q}=\gamma\varepsilon^*q^+ \,, \tag{4.1}$$

$$\dot{q}^+=\gamma\varepsilon q \,. \tag{4.2}$$

[10]Recently it has been shown that antibunching-type correlations are produced also in a laser field that undergoes spontaneous degenerate hyper-Raman scattering (Peřinová *et al.*, 1979b; Szlachetka *et al.*, 1980). Moreover, it has been pointed out by Yuen and Shapiro (1979) that degenerate four-wave mixing is also suited to generate antibunching properties in field modes that are proper combinations of the output object and image waves.

[11]The references mentioned are concerned with nondegenerate parametric interaction. The specialization to the degenerate case is a trivial matter.

Here q^+, q are the photon creation and annihilation operators, respectively, for the fundamental wave. The effective coupling constant γ, taken as positive, is proportional to both the nonlinear susceptibility of the medium and the amplitude of the harmonic. The phase factor ε is defined by the phase $\varphi_{2\omega}$ of the harmonic through the relation

$$\varepsilon=\exp[i(\varphi_{2\omega}-\tfrac{1}{2}\pi)] . \tag{4.3}$$

Differentiating Eq. (4.1) once more and utilizing Eq. (4.2), we find the second-order differential equation

$$\ddot{q}=\gamma^2 q , \tag{4.4}$$

which is easily solved to yield

$$q(t)=c(t)q^0+\varepsilon^* s(t)q^{+0} . \tag{4.5}$$

Here, the following abbreviations have been introduced

$$c(t)=\cosh(\gamma t), \quad s(t)=\sinh(\gamma t) , \tag{4.6}$$

and the upperscript zero refers to the initial state at $t=0$.

With this solution in hand, it is a straightforward matter to calculate the photon statistical properties of the fundamental wave as a function of time. After some algebra one obtains

$$q^+(t)q(t)=c^2q^{+0}q^0+\varepsilon cs(q^0)^2+\varepsilon^* cs(q^{+0})^2+s^2(1+q^{+0}q^0) , \tag{4.7}$$

$$\begin{aligned} q^{+2}(t)q^2(t)=&\,c^4(q^{+0})^2(q^0)^2+c^2s^2[4(q^{+0})^2(q^0)^2+8q^{+0}q^0+1]+s^4[(q^{+0})^2(q^0)^2+4q^{+0}q^0+2]\\ &+\varepsilon c^3s[2q^{+0}(q^0)^3+(q^0)^2]+\varepsilon cs^3[2q^{+0}(q^0)^3+5(q^0)^2]\\ &+\varepsilon^* c^3s[2(q^{+0})^3q^0+(q^{+0})^2]+\varepsilon^* cs^3[2(q^{+0})^3q^0+5(q^{+0})^2]\\ &+\varepsilon^2c^2s^2(q^0)^4+\varepsilon^{*2}c^2s^2(q^{+0})^4 . \end{aligned} \tag{4.8}$$

Supposing now the fundamental wave to be initially in a Glauber state $|\alpha\rangle$, we find the relevant physical quantity

$$\Delta\equiv\langle q^{+2}q^2\rangle-\langle q^+q\rangle^2 , \tag{4.9}$$

whose sign indicates whether bunching (+) or antibunching (−) occurs [see Eq. (2.39)], to be given by

$$\Delta(t)=A(t)+\bar{n}_0[B(t)+C(t)\cos\theta] , \tag{4.10}$$

where

$$\begin{aligned} A(t)&=c^2s^2+s^4\\ &=\tfrac{1}{4}[\cosh(4\gamma t)-2\cosh(2\gamma t)+1] ,\\ B(t)&=6c^2s^2+2s^4\\ &=\cosh(4\gamma t)-\cosh(2\gamma t) ,\\ C(t)&=2c^3s+6cs^3\\ &=\sinh(4\gamma t)-\sinh(2\gamma t) . \end{aligned} \tag{4.11}$$

The angle θ is defined as

$$\theta=\varphi_{2\omega}-2\varphi_\omega-\frac{\pi}{2} , \tag{4.12}$$

where φ_ω is the initial phase of the fundamental wave, i.e., the phase of the complex number α^*, and $\bar{n}_0$ is the mean photon number at $t=0$, $\bar{n}_0=|\alpha|^2$.

The first term on the right-hand side of Eq. (4.10), being independent of the incident fundamental wave, has its origin in spontaneous processes. In fact, it is well known that photons from an intense wave split, via parametric fluorescence, into photon pairs at lower frequencies. Specifically, in this way the harmonic gives rise to the emission of photon pairs into the fundamental wave. This spontaneously produced radiation becomes amplified in the course of interaction, and the first term in Eq. (4.10) represents precisely this amplified noise. In accordance with the bunching character of the latter, the term in question is always positive for $t>0$.

The second term in Eq. (4.10), on the contrary, may take negative values and even overcompensate the first one. In that case the fundamental wave will display the antibunching phenomenon. Equation (4.10) indicates that the antibunching effect will be most pronounced for $\theta=\pi$. Then the fundamental wave experiences maximum attenuation. This becomes evident from the expression for the mean photon number in the fundamental wave

$$\bar{n}(t)\equiv\langle q^+(t)q(t)\rangle=\bar{n}_0(c^2+s^2+2cs\cos\theta)+s^2 , \tag{4.13}$$

which follows from Eq. (4.7) under the above assumption that the fundamental wave is initially in a Glauber state.

For a quantitative estimation of the relative magnitude of the antibunching effect we consider the ratio $\Delta(t)/\bar{n}(t)$, which, at best, equals -1 [see Eq. (2.24) and the text subsequent to Eq. (2.39)].

When $\bar{n}_0$ is large in comparison to unity, the contributions from the spontaneous processes will become noticeable only after the fundamental wave has experienced strong damping. It follows from Eqs. (4.10) and (4.13) that in these circumstances the quantity $\Delta(t)/\bar{n}(t)$ will only weakly depend on $\bar{n}_0$ in the early stage of the interaction process. This becomes obvious also from Fig. 6, where the temporal variation of $\Delta(t)/\bar{n}(t)$ in the case $\theta=\pi$ is plotted for different values of $\bar{n}_0$. One recognizes that antibunching properties are in fact produced in the course of interaction. However, at a certain instant, which is attained the earlier the smaller the $\bar{n}_0$, antibunching is converted into bunching. Hence the anti-

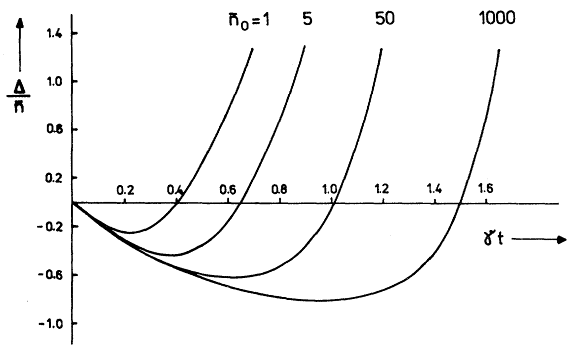

FIG. 6. The quantity $\Delta/\bar{n} \equiv (\Delta n^2 - \bar{n})/\bar{n}$ characteristic of the relative magnitude of the antibunching effect in the fundamental wave, vs time for different values of the initial mean photon number $\bar{n}_0$ as a parameter. (γ is the effective coupling constant.)

bunching phenomenon is a transient effect in parametric interaction, in contrast to the situation envisaged in multiphoton absorption.

Physically, the qualitative change in the photon statistical properties of the fundamental wave in the degenerate parametric process is easily understood from the fact that in the course of interaction the coherent part of this wave, which is a remnant of the initial state, becomes more and more attenuated. Hence the noise contribution, which is permanently amplified, will dominate, after sufficient time has elapsed, the length of this time interval increasing with growing $\bar{n}_0$.

One learns from Fig. 6 that the greater the $\bar{n}_0$, the longer the time needed to reach the respective minimum of $\Delta/\bar{n}$. Optimum conditions for the measurement of the antibunching effect at given $\bar{n}_0$, however, correspond to the minimum of $\Delta/\bar{n}^2$. Due to the decrease of the mean photon number in time, the positions of those minima are shifted towards larger values of γt, in comparison to the curves representing $\Delta/\bar{n}$. Numerical results concerning the minima of $\Delta/\bar{n}^2$ are listed in Table I. Obviously, the choice $\bar{n}_0=5$, $\gamma t=0.44$, for instance, offers good prospects for an actual observation of antibunching properties.

Since the time scale is determined by the inverse of the coupling constant γ, which is proportional to the amplitude of the harmonic, one can in fact adjust γ such that, given the length of the nonlinear crystal, the field exhibits maximum antibunching properties when leaving the crystal. Actually, the possibility of varying and, in particular, of enhancing the coupling constant via the intensity of the harmonic is a specific feature by which the parametric interaction process differs favorably from multiphoton absorption.

B. The nondegenerate case

In nondegenerate parametric interaction we are dealing, in conventional terminology, with a signal wave at frequency ω_s, an idler wave at ω_i, and a pump wave at $\omega_p=\omega_s+\omega_i$. As before, we assume the phase-matching condition to be fulfilled and the phases of the three waves to be adjusted such that both the signal and the idler wave are attenuated, whereas the intense pump wave (in contrast to what the name suggests!) becomes amplified.

Since the basic event, namely, the fusion of a signal and an idler photon into a pump photon, has the character of one-photon absorption for the signal and the idler wave, respectively, we do not expect those waves to acquire antibunching properties in the course of interaction. The two photons, however, are simultaneously removed from the respective waves; hence the intensities in the signal and the idler wave will be correlated. As has been shown by Trung and Schütte (1978) and by Peřinová and Peřina (1978), it is just anticorrelations that are generated between the two waves in the early stage of interaction. In the following, a straightforward treatment of this effect will be presented.

Similar to the degenerate case, we consider the pump wave as a classical field which, practically, remains unaffected by the interaction process. Then the equations of motion read [see, for example, Brunner and Paul (1977)]

$$\dot{q}_s = \gamma\varepsilon^* q_i^+ \ , \tag{4.14}$$

$$\dot{q}_i = \gamma\varepsilon^* q_s^+ \ , \tag{4.15}$$

where ε is given by Eq. (4.3) with $\varphi_{2\omega}$ replaced by φ_p.

TABLE I. Minimum values of $\Delta/\bar{n}^2$ attainable in degenerate parametric three-wave interaction for different values of the initial photon number in the fundamental wave $\bar{n}_0$. In addition, the corresponding values for γt (product of the coupling constant and the interaction time), the mean photon number $\bar{n}$, and the relative attenuation are listed.

$\bar{n}_0$	γt	$\bar{n}$	$\frac{\Delta}{\bar{n}^2}$	$1-\frac{\bar{n}}{\bar{n}_0}$
1	0.24	0.68	−0.38	32%
2.5	0.35	1.37	−0.25	45%
5	0.44	2.28	−0.18	54%
10	0.54	3.72	−0.12	63%
50	0.78	11.25	−0.051	78%
1000	1.29	78.59	−0.0083	92%

The solutions to Eqs. (4.14) and (4.15) are

$$q_s(t)=c(t)q_s^0+\varepsilon^* s(t)q_i^{+0}, \tag{4.16}$$

$$q_i(t)=c(t)q_i^0+\varepsilon^* s(t)q_s^{+0}. \tag{4.17}$$

Assuming the signal and the idler wave to be initially in Glauber states $|\alpha_s\rangle$ and $|\alpha_i\rangle$, respectively, one finds from Eqs. (4.16) and (4.17) after some algebra (Paul and Brunner, 1981)

$$\bar{n}_s(t)\equiv\langle q_s^+(t)q_s(t)\rangle =c^2|\alpha_s|^2+s^2(|\alpha_i|^2+1) +2cs|\alpha_s|\,|\alpha_i|\cos\theta, \tag{4.18}$$

$$\Delta_s(t)\equiv\langle q_s^{+2}(t)q_s^2(t)\rangle-\langle q_s^+(t)q_s(t)\rangle^2 =2s^2\{c^2|\alpha_s|^2 +2cs|\alpha_s|\,|\alpha_i|\cos\theta+s^2|\alpha_1|^2\}+s^4, \tag{4.19}$$

where

$$\theta=\varphi_p-\varphi_s-\varphi_i-\frac{\pi}{2} \tag{4.20}$$

(φ_s and φ_i being the phases of α_s^* and α_i^*, respectively). Similar relations hold for the idler wave, of course.

The quantity characteristic of cross correlations, on the other hand, becomes

$$\Delta_{\text{cross}}(t)\equiv\langle q_s^+(t)q_i^+(t)q_i(t)q_s(t)\rangle -\langle q_s^+(t)q_s(t)\rangle\langle q_i^+(t)q_i(t)\rangle =2c^2s^2(|\alpha_s|^2+|\alpha_i|^2+\tfrac{1}{2}) +2(c^3s+cs^3)|\alpha_s||\alpha_i|\cos\theta. \tag{4.21}$$

Since the bracket on the right-hand side of Eq. (4.19) is positive for $t>0$, the signal wave (and likewise the idler wave), when investigated alone, displays bunching properties, as we expected.

However, Δ_{cross} may take negative values—i.e., anticorrelations between the two beams may be generated. Obviously, the specification $\theta=\pi$ will provide the best opportunity to achieve this. This choice corresponds to maximum attenuation of both the signal and the idler wave, as is evident from Eq. (4.18). Let us assume, for the sake of simplicity, that the initial mean photon numbers are the same for the signal and the idler wave, $|\alpha_s|^2=|\alpha_i|^2$. Since the expression for $\bar{n}_i(t)$ differs from Eq. (4.18) only in that the subscripts s and i are interchanged, this remains so for all times, $\bar{n}_i(t)=\bar{n}_s(t)$. Moreover, also the fluctuations are equal in both beams $\Delta_s(t)=\Delta_i(t)$. In those circumstances, the quantity $\Delta_{\text{cross}}(t)$ for $\theta=\pi$ is indeed negative for not too large values of γt—i.e., the signal and the idler wave are anticorrelated. This becomes obvious from Fig. 7, in which the ratio $\Delta_{\text{cross}}(t)/\bar{n}_s(t)$ has been plotted versus time for different values of $\bar{n}_s(0)$.

As in the degenerate case, the contributions due to spontaneous processes are of less importance in the early

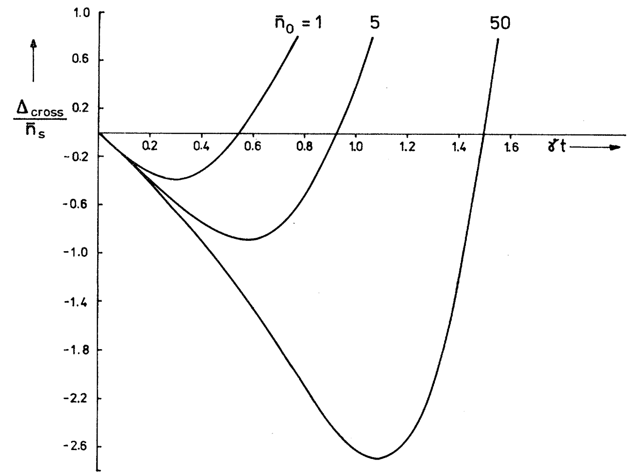

FIG. 7. The quantity $\Delta_{\text{cross}}/\bar{n}_s\equiv(\langle q_s^+q_i^+q_iq_s\rangle-\bar{n}_s\bar{n}_i)/\bar{n}_s$ that indicates the presence of cross correlations between the signal and the idler wave, vs time for different values of the initial mean photon number in the signal wave $\bar{n}_s(0)$, in the special case $\bar{n}_i(0)=\bar{n}_s(0)$.

stage of the interaction process which makes the ratio $\Delta_{\text{cross}}(t)/\bar{n}_s(t)$ only weakly depend on $\bar{n}_s(0)$, for $\bar{n}_s(0)\gg1$, near $\gamma t=0$, whereas they become dominant after a certain time has elapsed that increases with growing $\bar{n}_s(0)$, giving rise to a change of anticorrelations into correlations.

Of course, the occurrence of anticorrelations between two waves is no matter of surprise in classical wave theory. For example, anticorrelations have recently been observed in light scattering from nonspherical particles in dilute solution by measuring the cross correlation of signals from two spatially separated detectors, each of which received light from the same small number of scatterers (Griffin and Pusey, 1979).

Denoting the (fluctuating) deviation of the intensity in the beam 1 or 2, respectively, from its mean value by

$$i_1=I_1-\bar{I}_1,\quad i_2=I_2-\bar{I}_2, \tag{4.22}$$

we can write the classical analog of the quantity (4.21) (the cross-variance function) as

$$\Delta_{\text{cross}}^{\text{class}}(t)=\langle\langle i_1(t)i_2(t)\rangle\rangle, \tag{4.23}$$

where the double bracket denotes ensemble averaging.

Applying Schwarz's inequality, one obtains from Eq. (4.23) the following bound for the modulus of $\Delta_{\text{cross}}^{\text{class}}$, which is well known in classical (statistical) communication theory [cf., for instance, Middleton (1960)]

$$|\Delta_{\text{cross}}^{\text{class}}|\le(\Delta_1^{\text{class}}\Delta_2^{\text{class}})^{1/2}, \tag{4.24}$$

where $\Delta_\mu^{\text{class}}=\langle\langle i_\mu^2\rangle\rangle$ $(\mu=1,2)$ are the autovariances.

Now, what is really exciting from the classical point of view is the fact that the anticorrelations produced in nondegenerate parametric three-wave interaction are stronger in the early stage of the interaction process than those allowed in the framework of a classical description,—i.e., they violate the inequality

$$|\Delta_{\rm cross}| \le \Delta_s \tag{4.25}$$

corresponding to (4.24) in the special case $\Delta_i=\Delta_s$. This feature is displayed in Fig. 8, where both the quantities $-\Delta_{\rm cross}/\bar{n}_s$ and $\Delta_s/\bar{n}_s$ are plotted for $\bar{n}_s(0)=\bar{n}_i(0)=5$. Hence, the anticorrelations under discussion are in fact of nonclassical character.

The nonclassical behavior becomes also apparent in an antibunching effect exhibited by the superposition field made up by the signal and the idler wave. Indeed, the relative excess coincidence counting rate, in case the superposition field falls on both detectors in the Brown-and-Twiss—type setup and the detectors do not distinguish between the signal and the idler wave, takes the simple form (Paul and Brunner, 1981)

$$r(t)=\frac{\Delta_s(t)+\Delta_{\rm cross}(t)}{2\bar{n}_s(t)^2} \tag{4.26}$$

[for $\bar{n}_i(0)=\bar{n}_s(0)$], which clearly indicates that it is the "abnormal" strength of the anticorrelations giving rise to a violation of the classical inequality (4.25) that is responsible for the antibunching phenomenon.

Strictly speaking, in order to derive Eq. (4.26), the assumption $|\omega_s-\omega_i|\tau_{\rm resp}\gtrsim 1$ (ω_s,ω_i signal and idler frequencies, respectively, and $\tau_{\rm resp}$ response time of the detectors) had to be made. Physically, this means that the detectors average out the short-time fluctuations due to the beating of the two modes, which otherwise give rise to strong bunching in the superposition field. On the other hand, it has recently been shown by Bandilla and Ritze (1980b) that the above restriction can be avoided when the signal and the idler wave are linearly polarized in mutually orthogonal directions, their frequencies, however, being equal. Then the antibunching effect is displayed by the interference field produced by passing the two beams through an appropriately oriented analyzer (cf. also Sec. V.B).

Making a quantitative comparison with the degenerate process, one observes that Eq. (4.26), at given total number of photons in the initial state, $\bar{n}_s(0)+\bar{n}_i(0)=2\bar{n}_s(0)=\bar{n}_0$, differs from the corresponding expression for the degenerate case only in that the noise contributions are larger by a factor of 2. On the other hand, the quantity (4.26) is easily proved to be half the corresponding value for the relative excess coincidence counting rate in the degenerate case, provided $\bar{n}_0$ is set equal to $\bar{n}_s(0)=\bar{n}_i(0)$. Hence we infer from Table I in Sec. IV.A that for $\bar{n}_s(0)=\bar{n}_i(0)=2.5$ the optimum value of $r(t)$ is -0.12, appearing at $\gamma t=0.35$.

The antibunching properties of the superposition field have been extensively analyzed by Mišta and Peřina (1978) via a calculation of the characteristic function in the Fokker Planck description. Moreover, they included lossy mechanisms for both the signal and the idler wave in their treatment.

Finally, I should like to mention that anticorrelations have also been studied theoretically in two-photon absorption from two different beams (Simaan and Loudon,

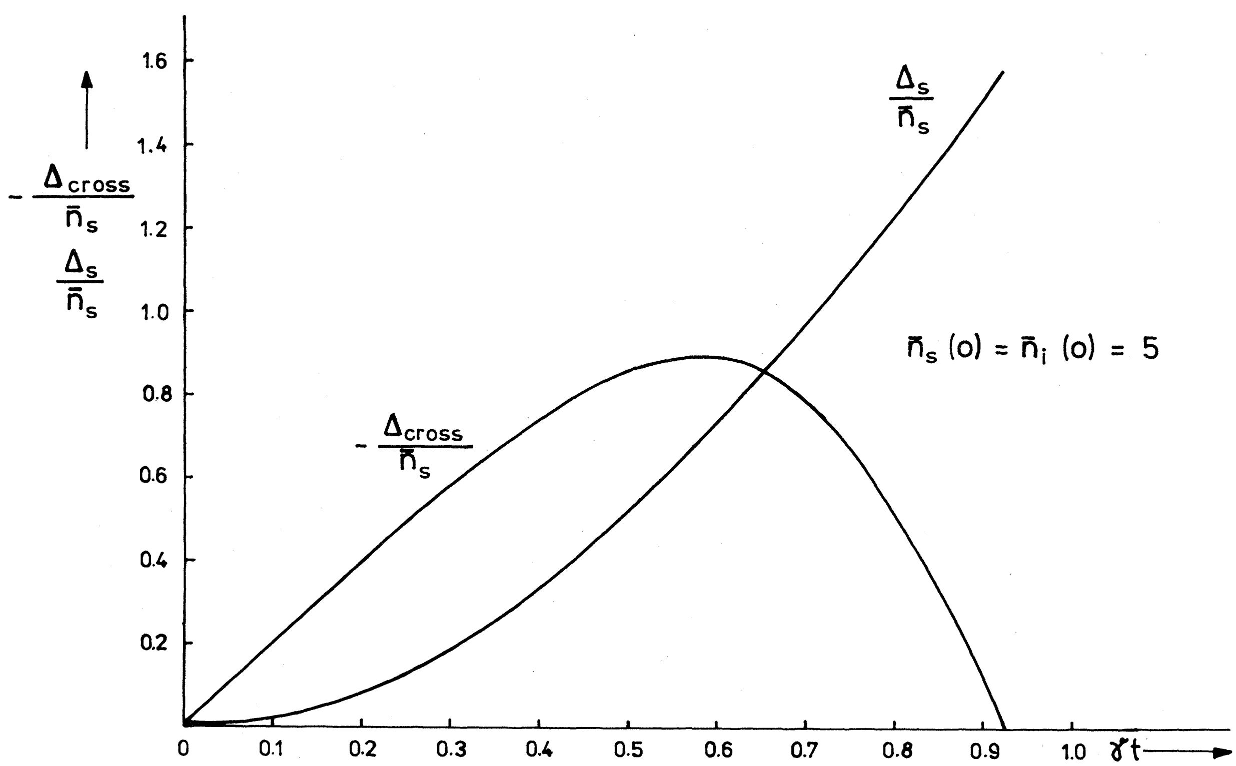

FIG. 8. Comparison between cross and autocorrelations in nondegenerate parametric three-wave interaction. $\Delta_{\rm cross}\equiv\langle q_s^+q_i^+q_iq_s\rangle-\bar{n}_s^2$ [for $\bar{n}_i(0)=\bar{n}_s(0)$] and $\Delta_s\equiv\Delta n_s^2-\bar{n}_s$.

1975b). Moreover, they have been shown to be generated also in both Raman and hyper-Raman scattering (Szlachetka, Kielich, Peřina, and Peřinová, 1979; Peřinová, Peřina, Szlachetka, and Kielich, 1979a, b; Szlachetka and Kielich, 1980; Szlachetka, Kielich, Peřina, and Peřinová, 1980; Tänzler and Schütte, 1981; Germey, Schütte, and Tiebel, 1981).

C. The mode volume

Having thus shown parametric three-wave interaction to be well suited to produce fields with antibunching properties, we are left with the problem of specifying the longitudinal dimension of the mode volume. Similar to the case of multiphoton absorption discussed in Sec. III.F, the task is to determine the critical value for the time delay τ_{crit} between two photons (in the degenerate case, these are two photons in the ground wave, and in the nondegenerate case a signal and an idler photon) which must not be exceeded by them in order to have a chance to undergo "fusion" into a harmonic (or pump) photon. Unlike the situation envisaged in multiphoton absorption, however, the nonlinear material involved in the parametric process does not provide a memory mechanism defining τ_{crit}, since the relaxation times are extremely short, so that the interaction has simultaneous character at a given position. However, a specific mechanism which effectively produces a nonlocal coupling either of the fundamental wave with itself or between the signal and the idler wave, has been found to originate from the (linear) dispersion of the nonlinear material (Paul and Brunner, 1980).

Let us first consider the nondegenerate case. Owing to the dispersion, the signal and the idler wave propagate with different group velocities v_s and v_i. This has the consequence that a field amplitude in the signal wave which begins to travel through the crystal—say, at an instant t—during its passage experiences the effect of not only the field amplitude in the idler wave which started at the same time, but also of idler amplitudes with starting times $t'<t$ (for $v_s>v_i$) or $t'>t$ (for $v_s<v_i$). The maximum value for $|t'-t|$ is given by the difference of the transit times for the signal and the idler wave, respectively,

$$\tau_{\text{crit}}=L\left|\frac{1}{v_s}-\frac{1}{v_i}\right| , \tag{4.27}$$

L being the crystal length.

When the derivative of the index of refraction μ with respect to ω varies slowly in the frequency range ω_s to ω_i, we may write

$$\tau_{\text{crit}}\approx\frac{2T}{\mu}\left|(\omega_s-\omega_i)\frac{\partial\mu}{\partial\omega}\right| \tag{4.28}$$

(nondegenerate case), where $T=L/v_s$ is the transit time.

In fact, the characteristic time (4.27) is nothing other than the inverse of the linewidth as determined by the phase-matching condition [see Kleinman (1968)]. This linewidth becomes manifest in the spontaneous process (parametric fluorescence); on the other hand, it characterizes that part of the spectrum of a broad-band signal (and, likewise, idler) wave that is actually affected in the parametric interaction.

In the degenerate case the difference of the group velocities at $\omega+\frac{1}{2}\Delta\omega$ and $\omega-\frac{1}{2}\Delta\omega$, instead of ω_s and ω_i, comes into play, where ω is the center frequency and $\Delta\omega$ the bandwidth of the fundamental wave. (The pump wave is still assumed to be monochromatic.) The corresponding difference in the transit times is

$$\tau=L\left|\frac{1}{v\left[\omega+\dfrac{\Delta\omega}{2}\right]}-\frac{1}{v\left[\omega-\dfrac{\Delta\omega}{2}\right]}\right|=L\,\Delta\omega\left|\frac{\partial^2 k}{\partial\omega^2}\right|$$
$$=L\frac{\Delta\omega}{v^3}\left|\frac{\partial v}{\partial k}\right| \quad \text{(degenerate case)} . \tag{4.29}$$

Assuming now the bandwidth $\Delta\omega$ to coincide with that allowed by the phase-matching condition, we can equate[12] τ and $2\pi/\Delta\omega$. This gives us the value for τ which can occur, at maximum, in degenerate parametric interaction

$$\tau_{\text{crit}}=2\left[\frac{\pi T}{\mu}\left|\frac{\partial\mu}{\partial\omega}\right|\right]^{1/2} \quad \text{(degenerate case)} , \tag{4.30}$$

T being the transit time as before.

Equations (4.28) and (4.30) indicate the time scale on which the antibunching (or anticorrelation) phenomenon, as predicted in the preceding sections in the single-mode formalism, will actually show up. Accordingly, the longitudinal dimension of the mode volume is given by $l_{\text{crit}}=c\tau_{\text{crit}}$.

In practice, the critical time τ_{crit} proves to be extremely short. In fact, it follows from Eqs. (4.28) and (4.30) for $|\partial\mu/\partial\omega|\approx10^{-17}$ s, $L=3$ cm, and $\omega_s-\omega_i\approx10^{15}$ Hz that τ_{crit} is of the order of a few picoseconds for the nondegenerate case, while it is smaller still, by a factor of 10 or so, for the degenerate case.

Physically, it appears to be clear that the time constants (4.28) and (4.30) are characteristic also of the process which is inverse to that considered so far, namely, the decay of a pump photon into both a signal and an idler photon (parametric fluorescence). Then, τ_{crit} describes the maximum time delay between the two photons generated in the same elementary process. This problem has been extensively studied by Mollow (1973).

V. ANTIBUNCHING AND INTERFERENCE

A. Introductory remarks

In the preceding sections we have analyzed physical processes which make it possible, at least in principle, to

[12]In fact, replacing τ in this way, one obtains from Eq. (4.29) the formula for the linewidth in parametric fluorescence, as derived by Kleinman (1968) (apart from a factor of 2).

change the photon statistics of light such that initial bunching (or nonbunching corresponding to a Glauber state) is converted into antibunching. A different question we will be concerned with in what follows is how antibunching properties thus produced will eventually change in further interactions that the field may undergo. This problem is of some practical interest particularly in the case of two- (or multi-) photon absorbed light. In fact, only high-intensity fields are appreciably affected by multiphoton absorbers. Hence the mean photon number $\bar{n}_{\rm out}$ (per mode volume) in the outgoing field will normally be very large, which practically precludes an observation of the antibunching phenomenon, whose magnitude is of the order of $1/\bar{n}_{\rm out}$.

Therefore, it would be desirable to have a physical mechanism at hand that diminishes the field intensity by orders of magnitude, without, however, remarkably reducing the quantity $\Delta/\bar{n}\equiv(\langle q^{+2}q^2\rangle-\langle q^+q\rangle^2)/\langle q^+q\rangle$, which measures, if negative, the magnitude of the antibunching effect in comparison to the optimum value -1, corresponding to a perfectly sharp photon number.

First, we note that one-photon absorption (or any equivalent process like beam splitting or the "dilution" of the field due to spreading in space) certainly does not work in the way required. In fact, we saw in Sec. III.A that the relative excess coincidence counting rate $\Delta/\bar{n}^2$ is conserved in this process. This means the quantity $\Delta/\bar{n}$ varies as $\bar{n}/\bar{n}(0)$, where $\bar{n}(0)$ is the mean photon number at the beginning of the damping process, and hence becomes negligibly small for remarkable attenuation—say, $\bar{n}/\bar{n}(0)\lesssim 10^{-3}$—irrespective of its initial value. So we have to look for different mechanisms.

In classical electrodynamics, there exists a well-known method of reducing the mean intensity of a field whose amplitude contains a fluctuating part, without affecting the latter, thus indeed enhancing the relative intensity fluctuations. It simply consists of making the field under investigation interfere destructively with a nonfluctuating coherent wave oscillating at the same frequency with a properly chosen amplitude. We are thus led to the idea [first suggested by Steudel (1977)] that interference of a field exhibiting antibunching properties with a reference beam being in a Glauber state might provide a mechanism of reducing the intensity without appreciably affecting the relative magnitude of the antibunching effect, $\Delta/\bar{n}$. Similar success might be achieved by dividing the field in question into coherent parts and making them interfere (destructively). Both possibilities have been thoroughly investigated by Bandilla and Ritze (1979, 1980a). [See also Ritze and Bandilla (1979a).] In the following, I shall outline the main features of their analysis.

B. Enhancement of antibunching through destructive interference

We study the interference of a field possessing antibunching properties with a coherent reference beam. For the sake of mathematical convenience we idealize both waves as single-mode states of the field. Those two plane-wave—type modes labeled $\lambda=1$ and $\lambda=2$, are assumed to differ only by their directions of propagation.[13] However, this difference can be so small that the spatial variation of the interference pattern can be neglected in a given domain in space. Since we are dealing with expectation values for normally ordered field operators, no contributions from vacuum fluctuations will enter our calculations. Hence we need retain only the terms corresponding to $\lambda=1$ and $\lambda=2$ in Eqs. (2.26). Introducing the abbreviations

$$E_\lambda^{(+)}(\mathbf{r},t)=a_2(\mathbf{r},t)q_\lambda\ , \tag{5.1}$$

we thus can write

$$E^{(\pm)}(\mathbf{r},t)=E_1^{(\pm)}(\mathbf{r},t)+E_2^{(\pm)}(\mathbf{r},t)\ . \tag{5.2}$$

Since the two beams are generated by different sources, they are statistically independent. Formally, this means that expectation values for operator products related to both beams factorize. Assuming the reference beam $\lambda=2$ to be in a Glauber state $|\beta\rangle$, we find after some simple algebra the following results:

$$\langle E^{(-)}E^{(+)}\rangle=|a|^2[\langle q_1^+q_1\rangle+2\,\mathrm{Re}(\langle q_1^+\rangle\beta)+|\beta|^2]\ , \tag{5.3}$$

$$\begin{aligned}\langle E^{(-)2}E^{(+)2}\rangle-\langle E^{(-)}E^{(+)}\rangle^2=|a|^4\{&\langle q_1^{+2}q_1^2\rangle-\langle q_1^+q_1\rangle^2+4\,\mathrm{Re}[(\langle q_1^{+2}q_1\rangle-\langle q_1^+\rangle\langle q_1^+q_1\rangle)\beta]\\&+2\,\mathrm{Re}[(\langle q_1^{+2}\rangle-\langle q_1^+\rangle^2)\beta^2]+2(\langle q_1^+q_1\rangle-\langle q_1^+\rangle\langle q_1\rangle)|\beta|^2\}\ .\end{aligned} \tag{5.4}$$

In accordance with the above assumption, we put $|a_1(\mathbf{r},t)|^2=|a_2(\mathbf{r},t)|^2=|a|^2$ (independent of $\mathbf{r}$ and t) and $a_1(\mathbf{r},t)a_2^*(\mathbf{r},t)\approx|a|^2$ in a given domain in space.

One learns from Eq. (5.4) that the photon statistical properties of the field resulting from interference are determined not only by the intensity fluctuations in the field $\lambda=1$, but also by the coherence properties of the latter which are reflected by the combinations of expectation values involved in the interference terms of Eq. (5.4). To evaluate those quantities in case of two-photon absorbed light is not at all a trivial matter. This problem has been treated by Simaan and Loudon (1978) and by Bandilla and Ritze (1980a), the latter paper being in fact devoted to a study of the interference effect with which we are presently dealing.

On the contrary, the degenerate parametric interaction

[13]The interference may be produced also with the help of polarizers. [For details see Ritze and Bandilla (1979a).]

process allows for a simple analytic solution in the Heisenberg picture [see Eq. (4.5)], with the help of which the expectation values in question are easily calculated. For this reason we confine our analysis to this case, the results being indeed similar to those obtained by Bandilla and Ritze (1980a) for two-photon absorbed light.

Under the assumption that the light in the fundamental wave traversing the nonlinear crystal is initially in a Glauber state $|\alpha\rangle$, where α is taken as a positive number for convenience, we find from Eq. (4.5) the quantities of interest to be

$$\langle q_1^{+2} q_1\rangle - \langle q_1^+\rangle\langle q_1^+ q_1\rangle = -s(c-s)^2\alpha \,, \tag{5.5}$$

$$\langle q_1^{+2}\rangle - \langle q_1^+\rangle^2 = -cs \,, \tag{5.6}$$

$$\langle q_1^+ q_1\rangle - \langle q_1^+\rangle\langle q_1\rangle = s^2 \,. \tag{5.7}$$

Here, we put $\varepsilon=-1$, thus specifying the phase of the pump wave such that the antibunching effect becomes maximum. As before, $c\equiv c(t_i)$ and $s\equiv s(t_i)$ are the abbreviations (4.6), where the duration of the interaction has been denoted by t_i in order to avoid confusion.

Insertion of the results (5.5)–(5.7) in Eq. (5.4) yields

$$\begin{aligned}\langle E^{(-)2}E^{(+)2}\rangle - \langle E^{(-)}E^{(+)}\rangle^2 &= |a|^4\{\Delta(t_i) - 4\,\mathrm{Re}[s(c-s)^2\alpha\beta] \\ &\quad -2\,\mathrm{Re}(cs\beta^2) + 2s^2|\beta|^2\} \quad (\alpha>0) \,,\end{aligned} \tag{5.8}$$

where Δ is given by Eq. (4.10) for $\cos\theta=-1$.

The mean intensity of the field produced by interference follows from Eq. (5.3) to be

$$\begin{aligned}\langle E^{(-)}E^{(+)}\rangle = |a|^2\{&(c-s)^2\alpha^2 \\ &+ s^2 + 2\,\mathrm{Re}[(c-s)\alpha\beta] + |\beta|^2\} \\ &(\alpha>0) \,,\end{aligned} \tag{5.9}$$

where the relations (4.13) and

$$\langle q^+\rangle = (c-s)\alpha \quad (\alpha>0) \tag{5.10}$$

have been used.

As mentioned above, we are interested in destructive interference. It becomes evident from Eq. (5.9) that the intensity of the total field will attain its minimum value (in the spatial domain considered) when $\beta=-(c-s)\alpha$. Allowing for a deviation from this special value, we put

$$\beta = -(c-s)\alpha + \delta \,, \tag{5.11}$$

where δ is assumed to be a real quantity. A straightforward calculation then gives us the quantity which characterizes the relative magnitude of the antibunching effect for the total field (in comparison to the optimum value -1)

$$\frac{\langle E^{(-)2}E^{(+)2}\rangle - \langle E^{(-)}E^{(+)}\rangle^2}{|a|^2\langle E^{(-)}E^{(+)}\rangle} = \frac{s^2(c^2+s^2) + [(c-s)^2-1]\delta^2}{\delta^2+s^2} \tag{5.12}$$

[cf. Ritze and Bandilla (1979a)].

Let us consider two special cases.

(i) The field intensity is reduced to its minimum value; then δ equals zero and Eq. (5.12) becomes

$$\begin{aligned}\frac{\langle E^{(-)2}E^{(+)2}\rangle - \langle E^{(-)}E^{(+)}\rangle^2}{|a|^2\langle E^{(-)}E^{(+)}\rangle} &= c^2+s^2 \\ &= 1+2s^2 \,.\end{aligned} \tag{5.13}$$

Since the corresponding value for thermal (chaotic) light is unity, Eq. (5.13) indicates "superbunching" to occur, rather than antibunching.

(ii) The parameter δ is chosen such that both the inequalities

$$\delta^2 \gg s^2 \tag{5.14}$$

and

$$\delta^2 \gg \left|\frac{s^2(c^2+s^2)}{(c-s)^2-1}\right| \tag{5.15}$$

are fulfilled for given t_i. [For example, when $\gamma t_i=0.4$, the right-hand sides in (5.14) and (5.15) are 0.17 and 0.41, respectively.] Then the ratio (5.12) reduces to the simple form

$$\begin{aligned}&\frac{\langle E^{(-)2}E^{(+)2}\rangle - \langle E^{(-)}E^{(+)}\rangle^2}{|a|^2\langle E^{(-)}E^{(+)}\rangle} \\ &\quad \approx e^{-2\gamma t_i} - 1 \quad (=-0.55 \text{ for } \gamma t_i=0.4) \,.\end{aligned} \tag{5.16}$$

Thus it has been demonstrated that the relative magnitude of the antibunching effect, in the above-mentioned sense, is essentially preserved in destructive interference under properly chosen experimental conditions. Since, on the other hand, the mean intensity is drastically reduced in case of large values of $\langle q_1^+ q_1\rangle$, the absolute magnitude of the antibunching effect, as given by the relative excess coincidence counting rate (2.32), is enhanced in the same proportion.

Some remarks are appropriate. First, a similar enhancement of the antibunching effect is accomplished also by "interference of the photon with itself," i.e., by a conventional interference experiment, where the initial light beam (which, in our case, is assumed to display the antibunching phenomenon) is divided into two coherent parts which are made to interfere. This has been shown by Bandilla and Ritze (1979).

Second, it is also possible to produce low-intensity fields with pronounced antibunching properties by destructive interference of the signal and the idler wave in nondegenerate parametric three-wave interaction (signal and idler fields differing in their directions of polarization, but not in their frequencies), as has been pointed out by Bandilla and Ritze (1980b).

Finally, one observes that the amplitude α of the fundamental wave entering the nonlinear crystal has completely disappeared from formula (5.12). This implies α might as well be equal to zero. In this case, however, the field emerging from the nonlinear crystal is nothing else than amplified noise (due to parametric fluorescence—see Sec. IV.A), and hence certainly has no antibunching properties. Nevertheless, the interference with a properly chosen reference beam produces a field that exhibits the antibunching effect. As pointed out by Ritze and Bandilla (1979a), an explanation of this really striking result can be found only in the fact that parametric noise, in contrast to chaotic light generated by spontaneous emission from excited atoms, has well-defined coherence properties impressed on it by the coherent pump wave.

Specifically, the sum of the phases of the signal and the idler wave, respectively, which have been been initiated by parametric fluorescence, is strongly correlated with the phase of the (coherent) pump wave [see, for example, Paul (1973)]. For the degenerate parametric process this implies that the phase of the amplified noise at the fundamental frequency is well defined in relation to the phase of the incident harmonic.

C. Transformation of phase fluctuations into intensity fluctuations

The observation made at the end of the foregoing section that coherence properties of a field will become manifest in the photon statistics when the field is made to interfere with an ideal reference beam, is of course no matter of surprise in classical electrodynamics, when considered from a general point of view, i.e., disregarding the fact that it is just antibunching properties that are acquired by the interference field in the above case. In fact, it is well known that the position of interference fringes is determined by the relative phase between the two interfering beams. Hence when one of them undergoes phase fluctuations, the intensity of the interference field at a given point in space will exhibit intensity fluctuations.

While in classical optics such a mechanism will certainly always lead to bunching, one might suspect that the quantum-mechanical formalism, in specific circumstances, allows antibunching properties, also, to be generated in this way. Indeed, Ritze and Bandilla (1979b) succeeded in proving that phase fluctuations that are produced by transmitting the light through a (transparent) medium with a nonlinear index of refraction can be transformed in antibunching-type intensity fluctuations through interference. In the following we will be concerned with this problem.

As has been pointed out by Eimerl (1978), in the case of a medium with a nonlinear index of refraction, the energy levels E_n of a particular mode of the radiation field are no longer equally spaced; instead, the level difference becomes dependent on the photon number n. Confining oneself to the lowest-order correction in n which corresponds to the Kerr effect, one can write

$$E_n = n\hbar\left[\omega_0 + \frac{n}{2}\Delta\omega\right] \quad (\Delta\omega \ll \omega_0) . \tag{5.17}$$

Due to the assumed reality of $\Delta\omega$, this formula introduced by Eimerl (1978) on the basis of a general argument, describes in fact a specific kind of phase fluctuations. Strictly speaking, the n-dependent corrections to the mode frequency ω_0 in Eq. (5.17) give rise to a dephasing, in the Schrödinger picture, of the coefficients c_n in a single-mode field state of the general form

$$\psi(t) = \sum_n c_n(t)\,|\,n\,\rangle ,$$
$$c_n(t) = c_n(0)e^{-iE_n t/\hbar} , \tag{5.18}$$

while the values for $|\,c_n\,|^2$ obviously retain their initial values. This means the photon statistics remain unchanged in the temporal evolution. However, a Glauber state will evolve into a state that differs from a Glauber state. This implies that the phase of the field is no longer so well defined as in a Glauber state—i.e., phase fluctuations are produced.

Formally, it is advantageous to rewrite Eq. (5.17) as

$$E_n = n\hbar\left[\omega' + \frac{n+1}{2}\Delta\omega\right] \quad (\omega' = \omega_0 - \tfrac{1}{2}\Delta\omega) . \tag{5.19}$$

This form of the eigenvalues corresponds to the Hamiltonian

$$H = H_0 + H_{\text{int}} , \tag{5.20}$$

where

$$H_0 = \hbar\omega' q^+ q \tag{5.21}$$

and

$$H_{\text{int}} = \tfrac{1}{2}\hbar\Delta\omega(q^+q)^2 + \tfrac{1}{2}\hbar\Delta\omega q^+ q . \tag{5.22}$$

In the interaction picture, the interaction Hamiltonian (5.22) leads to the following equation of motion for the photon creation operator:

$$\dot{q}^+ = i\Delta\omega q^+ q q^+ \tag{5.23}$$

[cf. also Ritze and Bandilla (1979b)].

Since q^+q is a constant of motion, the solution to Eq. (5.23) is simply given by

$$q^+(t) = e^{i\Delta\omega t q^{+0} q^0} q^{+0} , \tag{5.24}$$

where the upperscript zero refers to the initial state at $t=0$.

For an analysis of the photon statistical properties of the interference field we need the knowledge of the expectation values occurring on the right-hand side of Eqs. (5.3) and (5.4). As previously, we assume that the light beam which passes through the medium with a nonlinear index of refraction is initially in a Glauber state $|\,\alpha\rangle$. Then the expectation values of interest can be calculated from Eq. (5.24) in a straightforward manner. In the evaluation the following auxiliary formulas are useful:

$$e^{-xq^+q}q^+ = e^{-x}q^+e^{-xq^+q} \tag{5.25}$$

and

$$\langle \alpha | e^{-xq^+q} | \alpha \rangle = e^{(e^{-x}-1)|\alpha|^2} , \tag{5.26}$$

where x is a parameter. Both formulas are easily derived by means of the following lemma proved by Louisell (1973, p. 156)

$$e^{-xq^+q} = \sum_{l=0}^{\infty} \frac{(e^{-x}-1)^l}{l!} q^{+l} q^l . \tag{5.27}$$

After some algebra one obtains

$$\langle q^+(t) q(t) \rangle = |\alpha|^2 , \tag{5.28}$$

$$\langle q^+(t) \rangle = e^{i\Delta\omega t} e^{(e^{i\Delta\omega t}-1)|\alpha|^2} \alpha^* , \tag{5.29}$$

$$\langle q^{+2}(t) \rangle = e^{3i\Delta\omega t} e^{(e^{2i\Delta\omega t}-1)|\alpha|^2} \alpha^{*2} , \tag{5.30}$$

$$\langle q^{+2}(t) q(t) \rangle = e^{2i\Delta\omega t} e^{(e^{i\Delta\omega t}-1)|\alpha|^2} |\alpha|^2 \alpha^* . \tag{5.31}$$

With these results in hand, one finds from Eqs. (5.3) and (5.4) the mean intensity and the intensity correlations for the field produced by interference, with a reference beam being in a Glauber state $|\beta\rangle$, to be given by

$$\langle E^{(-)} E^{(+)} \rangle = |a|^2 [|\tilde{\alpha}|^2 + 2\,\mathrm{Re}(e^{(e^{i\varepsilon}-1)|\tilde{\alpha}|^2} \tilde{\alpha}^* \beta) + |\beta|^2] \tag{5.32}$$

and

$$\begin{aligned} \langle E^{(-)2} E^{(+)2} \rangle - \langle E^{(-)} E^{(+)} \rangle^2 = 2|a|^4 \{ & (1 - e^{(e^{i\varepsilon}-1)|\tilde{\alpha}|^2} e^{(e^{-i\varepsilon}-1)|\tilde{\alpha}|^2}) |\tilde{\alpha}|^2 |\beta|^2 \\ & + 2\,\mathrm{Re}[e^{(e^{i\varepsilon}-1)|\tilde{\alpha}|^2} (e^{i\varepsilon}-1) |\tilde{\alpha}|^2 \tilde{\alpha}^* \beta] \\ & + \mathrm{Re}[(e^{i\varepsilon} e^{(e^{2i\varepsilon}-1)|\tilde{\alpha}|^2} - e^{2(e^{i\varepsilon t}-1)|\tilde{\alpha}|^2}) \tilde{\alpha}^{*2} \beta^2] \} , \end{aligned} \tag{5.33}$$

where the following abbreviations have been introduced:

$$\tilde{\alpha} = \alpha e^{-i\varepsilon} , \tag{5.34}$$

$$\varepsilon = \Delta\omega t_i , \tag{5.35}$$

t_i being the duration of the interaction with the medium that possesses a nonlinear index of refraction.

Without loss of generality we can put

$$\tilde{\alpha} = -A e^{-i\phi} \quad (A > 0) , \qquad \beta = B > 0 . \tag{5.36}$$

Then Eqs. (5.32) and (5.33) can be written more compactly as [cf. Ritze and Bandilla (1979b)]

$$\langle E^{(-)} E^{(+)} \rangle = |a|^2 [A^2 - 2AB e^{(\cos\varepsilon - 1)A^2} \cos(A^2 \sin\varepsilon + \phi) + B^2] , \tag{5.37}$$

$$\begin{aligned} \langle E^{(-)2} E^{(+)2} \rangle - \langle E^{(-)} E^{(+)} \rangle^2 = 2|a|^4 A^2 B \{ & 2A e^{(\cos\varepsilon - 1)A^2} [\cos(A^2 \sin\varepsilon + \phi) - \cos(A^2 \sin\varepsilon + \phi + \varepsilon)] \\ & + B e^{[\cos(2\varepsilon)-1]A^2} \cos(A^2 \sin(2\varepsilon) + 2\phi + \varepsilon) \\ & - B e^{2(\cos\varepsilon - 1)A^2} [\cos(2A^2 \sin\varepsilon + 2\phi) + 1] + B \} . \end{aligned} \tag{5.38}$$

In order to show that this rather involved expression actually predicts antibunching to occur in certain circumstances, we specialize to the following case:

$$\begin{aligned} & \varepsilon \ll 1 , \\ & A^2 \sin\varepsilon + \phi \equiv \varphi = O(\varepsilon^{1/2}), \quad A^2 \varepsilon \equiv c = O(1) , \\ & A = (1+\rho) B, \quad \text{where } \rho = O(\varepsilon^{1/2}) . \end{aligned} \tag{5.39}$$

We then find by expansion [cf. Ritze and Bandilla (1979b)]

$$\langle E^{(-)} E^{(+)} \rangle = |a|^2 B^2 (\rho^2 + \varphi^2 + c\varepsilon) , \tag{5.40}$$

$$\langle E^{(-)2} E^{(+)2} \rangle - \langle E^{(-)} E^{(+)} \rangle^2 = 2|a|^4 c B^2 [2\varphi\rho + (c^2 + \tfrac{1}{2})\varepsilon + 2c\varphi^2] . \tag{5.41}$$

One learns from Eq. (5.41) that the resulting field displays the antibunching phenomenon when the phase φ and the relative difference in the initial amplitudes ρ are properly adjusted. Choosing, for example, the parameters as

$$\varphi = -\varepsilon^{1/2}, \quad \rho = 2\varepsilon^{1/2}, \quad c = \tfrac{1}{2} \quad (\varepsilon^{1/2} \ll 1) , \tag{5.42}$$

and observing the relation $\varepsilon B^2 \approx \varepsilon A^2 = c = \frac{1}{2}$, we obtain

$$\begin{aligned} & \langle E^{(-)} E^{(+)} \rangle = 2.75 |a|^2 , \\ & \langle E^{(-)2} E^{(+)2} \rangle - \langle E^{(-)} E^{(+)} \rangle^2 \approx -1.125 |a|^4 , \end{aligned} \tag{5.43}$$

from which the quantity characteristic of the relative magnitude of the antibunching effect follows to be

$$\frac{\langle E^{(-)2} E^{(+)2} \rangle - \langle E^{(-)} E^{(+)} \rangle^2}{|a|^2 \langle E^{(-)} E^{(+)} \rangle} \approx -0.41 . \tag{5.44}$$

This result indicates that it is indeed possible to transform the phase fluctuations acquired by a beam during its passage through a medium with a nonlinear index of refraction in antibunching-type intensity fluctuations of remarkable magnitude by destructive interference with an ideal reference beam. From the experimental point of view this technique has the additional advantage that the initial mean photon number $A^2=c/\varepsilon$ becomes drastically reduced in the process of interference.

It follows from Eq. (5.41) that the photon statistical properties of the interference field very sensitively depend on φ (and similarly, on ρ). In fact, putting, for example $\varphi=-\frac{1}{2}\varepsilon^{1/2}$, $\rho=\varepsilon^{1/2}$, $c=\frac{1}{2}$, we find $\langle E^{(-)2}E^{(+)2}\rangle-\langle E^{(-)}E^{(+)}\rangle^2=0$, while for $\varphi=0$ bunching occurs. In the special case $\rho=\varphi=0$, the mean intensity takes its minimum value, and the relative coincidence counting rate becomes

$$\frac{\langle E^{(-)2}E^{(+)2}\rangle-\langle E^{(-)}E^{(+)}\rangle^2}{\langle E^{(-)}E^{(+)}\rangle^2}=2+\frac{|a|^2}{\langle E^{(-)}E^{(+)}\rangle}\;. \tag{5.45}$$

Since the corresponding value for thermal radiation equals unity, Eq. (5.45) indicates the presence of "superbunching."

An experimental scheme similar to that considered in the foregoing, which utilizes the effect of self-induced gyrotropic birefringence, has been analyzed by Tanaś and Kielich (1979). Starting from a phenomenological interaction Hamiltonian that contains the hyperpolarizability tensors for the molecules, they investigated theoretically the interaction of an intense, elliptically polarized light beam with an isotropic medium. They showed that the outgoing light, after having passed an analyzer, exhibits either bunching or antibunching properties, dependent on the azimuth and the ellipticity of the incident beam (for given orientation of the analyzer). This study has been extended by Ritze (1980), who gave a microscopic description of the interaction and avoided the "short optical path approximation" used by Tanaś and Kielich (1979). Moreover, he pointed out that by insertion of a birefringent plate between the sample cell and the analyzer it becomes possible to produce a field that displays the antibunching effect at drastically reduced intensities, thus improving the prospects for an actual observation of this phenomenon.

It is interesting to note that the interaction of light with a medium that possesses a nonlinear index of refraction offers a novel technique of measuring the photon statistical properties of a light beam, as has been pointed out by Ritze and Bandilla (1979c). The procedure is as follows.[14] The initial beam whose photon statistical properties will be determined is transmitted through a Michelson-type interferometer with a Kerr cell inserted in one of its arms. The reflectivity of the entrance mirror dividing the incident beam into two coherent parts is $R=\cos^2\theta$. (For definiteness, assume that the transmitted partial beam passes through the nonlinear medium.) The arm lengths of the interferometer are to be adjusted such that the intensity of the outgoing field takes its minimum value. According to Ritze and Bandilla (1979c), the corresponding mean photon number is given by

$$\bar{n}_{\min}=\varepsilon^2\bar{N}^2\sin^4\theta\cos^2\theta\left[1+\left[\frac{\Delta N^2}{\bar{N}}-1\right]\sin^2\theta\right] \tag{5.46}$$

(ΔN^2 is the variance of the photon number and $\bar{N}$ is the mean photon number in the initial beam), provided the conditions $\bar{N}\gg 1$ and $(N-N)^3\ll\bar{N}^2$ are fulfilled.

It becomes obvious from Eq. (5.46) that the measurement of the minimum intensity, in dependence on the mirror reflectivity $R=\cos^2\theta$, suffices to determine the ratio $\Delta N^2/\bar{N}$. In this way, the photon statistical properties of the incident light can be measured without any coincidence counting technique being needed.

A variant of this detection scheme which makes use of self-induced gyrotropic birefringence has been discussed by Ritze (1980).

VI. RESONANCE FLUORESCENCE

A. Scattering by a single atom

In the preceding sections different interaction processes have been studied which have the common feature of being suited to change the photon statistics such as to endow fields that are initially in a coherent (Glauber) state with antibunching properties. Resonance fluorescence radiation emitted by a single atom, on the other hand, provides an opportunity to generate, from the very beginning, fields that exhibit the antibunching phenomenon, as has been predicted by Carmichael and Walls (1976a, b) and Kimble and Mandel (1976, 1977). [See also Cohen-Tannoudji (1977).] In the following I will present the main features of the theoretical analysis.

Basic to the theoretical description of resonance fluorescence is the complete determination of the scattered field by the atomic variables. This reflects the fact that what we call resonance fluorescence radiation is irradiated by an oscillating atomic dipole moment which itself is induced by the incident (coherent) field assumed

[14]The authors also consider a different scheme, where only one of the two perpendicularly polarized components of an initially linearly polarized light beam traversing the nonlinear medium feels a nonlinear index of refraction. (This is achieved by applying a transverse magnetic field.) Before falling on a detector, the light passes a Pockels cell and afterwards an analyzer which is precisely crossed with respect to the initial polarization direction. The Pockels cell is used to produce a phase difference between the two orthogonally polarized components which minimizes the intensity of the light transmitted through the analyzer. Formula (5.46) applies to this case, too, θ being now the angle between the magnetic field and the direction of transmission for the analyzer.

to be resonant with a specific atomic transition. In the quantum-mechanical formalism that electric field strength (in the far-field zone) is related to the raising and lowering operators a^+ and a, respectively, of an atom idealized as a two-level system, in the form (Mollow, 1969)

$$E^{(+)}(\mathbf{r},t)=f(\mathbf{r})a\left[t-\frac{r}{c}\right],$$

$$E^{(-)}(\mathbf{r},t)=f^*(\mathbf{r})a^+\left[t-\frac{r}{c}\right]. \qquad (6.1)$$

Here $f(\mathbf{r})$ is a well-known function that describes the spatial distribution of a classical dipole field. It is, however, of no interest to us, since the factors $f(\mathbf{r}),f^*(\mathbf{r})$ will cancel in forming the relative excess coincidence counting rate that characterizes the photon statistical properties of the field. Hence the problem of calculating correlation functions for the scattered field reduces to the determination of the corresponding atomic correlations.

From Eqs. (6.1), together with the familiar commutation relations for the atomic raising and lowering operators,

$$a^2=a^{+2}=0,\quad \{a,a^+\}\equiv aa^+ +a^+a=1\ , \qquad (6.2)$$

we immediately derive the fundamental result

$$\langle E^{(-)2}(\mathbf{r},t)E^{(+)2}(\mathbf{r},t)\rangle$$

$$=|f(\mathbf{r})|^4\left\langle a^{+2}\left[t-\frac{r}{c}\right]a^2\left[t-\frac{r}{c}\right]\right\rangle=0\ . \qquad (6.3)$$

This means the probability of simultaneously detecting two photons vanishes exactly—i.e., the atom emits one photon after the other, but never two photons at the same instant. In other words, the (nondelayed) coincidence counting rate is zero, while the individual counting rates of the detectors are certainly not. Hence the excess coincidence counting rate is negative, which, according to the criterion established in Sec. II.A, indicates antibunching to be present. Moreover, it immediately follows from the complete absence of nondelayed coincidences that the relative excess coincidence counting rate (2.9) equals -1, corresponding to a one-photon state, according to Eq. (2.24). Thus the principal capability of resonance fluorescence to generate fields with antibunching properties has already been demonstrated.

To get information on the correlation time of the antibunching effect thus produced, we follow an elegant treatment of Loudon (1980).

The atom experiences both a coherent driving force due to the incident field and the effect of radiation damping. The latter process can be accounted for, in an approximation based on a Markoff factorization assumption, by simply supplementing the Schrödinger equation for the atomic density operator ρ by phenomenological damping terms, as has been shown by Mollow and Miller (1969) [cf. also Mollow (1969)]. In the interaction representation, the equations of motion for the density matrix elements read [Allen and Eberly (1975); see also Mollow (1969)],

$$\frac{d\rho_{21}}{dt}=-\frac{\gamma}{2}\rho_{21}+ig\alpha(\rho_{22}-\rho_{11})\ , \qquad (6.4)$$

$$\frac{d\rho_{22}}{dt}=-\gamma\rho_{22}-ig\alpha\rho_{12}+ig^*\alpha^*\rho_{21}\ , \qquad (6.5)$$

the remaining matrix elements being determined by the relations

$$\rho_{12}=\rho_{21}^*,\quad \rho_{11}+\rho_{22}=1\ . \qquad (6.6)$$

Here, g is the coupling constant, α stands for the (complex) amplitude of the classical monochromatic driving field, and γ denotes the natural decay rate of the upper atomic level 2. For simplicity, we assume the incident field to be in exact resonance with the atomic transition. Moreover, we disregard additional damping mechanisms due to the environment of the atoms, e.g., collisions.

Now, we make use of the well-known fact that the expectation values for the operators a, a^+, and a^+a (in the interaction representation) are related to the ρ_{ik} in the following simple manner:

$$\langle a^+(t)\rangle=\rho_{12}(t),\quad \langle a(t)\rangle=\rho_{21}(t)\ , \qquad (6.7)$$

$$\langle a^+(t)a(t)\rangle=\rho_{22}(t)\ . \qquad (6.8)$$

From the linearity of the equations of motion (6.4) and (6.5) we conclude that any density matrix element $\rho_{ik}(t)$ depends *linearly* on the initial values $\rho_{12}(0)$, $\rho_{21}(0)$, and $\rho_{22}(0)$. [Note that by virtue of the second equation in (6.6), $\rho_{11}(0)$ can be expressed through $\rho_{22}(0)$.] Hence, observing the relationship (6.7) and (6.8), we are entitled to write, in particular,

$$\langle a^+(t)a(t)\rangle=\alpha_1(t)+\alpha_2(t)\langle a(0)\rangle+\alpha_3(t)\langle a^+(0)\rangle$$

$$+\alpha_4(t)\langle a^+(0)a(0)\rangle\ , \qquad (6.9)$$

where the functions $\alpha_j(t)$ $(j=1,2,3,4)$ are found by solving the equations of motion (6.4) and (6.5).

Due to the presence of a damping mechanism, the system will tend, after sufficient time has elapsed from the interaction being "switched on," to a steady state in which all information about the initial state is "forgotten." Formally, this means that the following relations hold:

$$\alpha_2(\infty)=\alpha_3(\infty)=\alpha_4(\infty)=0\ , \qquad (6.10)$$

$$\langle a^+a\rangle_{\text{st}}=\alpha_1(\infty)\ , \qquad (6.11)$$

where the subscript st has been used to indicate the steady-state value.

What we are ultimately interested in is not single-time expectation values, but two-time correlation functions. Fortunately, there exists a famous theorem, the so-called quantum regression theorem proved by Lax (1966, 1967) [cf. also Lax (1968)] for Markovian systems, which allows one to express any two-time correlation function through single-time expectation values. In explicit terms, this theorem states the following: if M is a member (or a linear combination) of a complete set of system Markovian operators M_μ, then the time evolution of the expectation value of M can be written as

$$\langle M(t)\rangle = \sum_\mu \beta_\mu(t)\langle M_\mu(0)\rangle \quad (t>0)\,, \tag{6.12}$$

and the mean of a two-time operator $L(t)M(t+\tau)N(t)$, where L and N are any system operators, is given by

$$\langle L(t)M(t+\tau)N(t)\rangle = \sum_\mu \beta_\mu(\tau)\langle L(t)M(t)N(t)\rangle\ . \tag{6.13}$$

Using this theorem to evaluate the steady-state value of the correlation function $\langle a^+(t)a^+(t+\tau)a(t+\tau)a(t)\rangle$, we find from Eq. (6.9) the simple result

$$\langle a^+(t)a^+(t+\tau)a(t+\tau)a(t)\rangle_{\rm st} = \alpha_1(\tau)\langle a^+a\rangle_{\rm st}\ . \tag{6.14}$$

[Note that the remaining terms in the sum on the right-hand side of Eq. (6.13) vanish in the present case by virtue of the commutation relations (6.2).]

It follows from Eq. (6.9), in combination with the relations (6.7) and (6.8), that $\alpha_1(\tau)$ has a simple physical interpretation: it describes the population of the upper level $\rho_{22}(0\,|\,\tau)$ at time τ following the turn-on of the interaction, when the atom starts in the lower state. In other words, $\alpha_1(\tau)$ is the conditional probability of finding the atom in the upper level at time $t=\tau$, given that it was in the lower level, with certainty, at $t=0$. On the other hand, the second factor on the right-hand side of Eq. (6.14) represents the steady-state value for the population of the upper level.

In view of Eqs. (6.1), (2.27), and (2.28), the delayed coincidence counting rate registered at a given point $\mathbf{r}$ is proportional to the expression (6.14). Hence the aforementioned physical interpretation of the two factors appearing on the right-hand side of Eq. (6.14) suggests the following picture for the emission process in resonance fluorescence, with regard to photon counting experiments: the emission of a photon is associated with an atomic transition from the upper to the lower level. Consequently, the probability of such an event happening is proportional to the mean population of the upper level $\rho_{22}^{\rm st} = \langle a^+a\rangle_{\rm st}$. Since the atom is in the lower state immediately after an emission of a photon has taken place, it is unable to emit a second photon at the same instant. Instead, it needs some "recovery time," during which the driving field brings it back to the upper level from which a new emission process may start. It is just this repopulation of the upper level that is described by the factor $\alpha_1(\tau)=\rho_{22}(0\,|\,\tau)$ in Eq. (6.14).

Equations (6.1) imply the mean intensity of the field—and hence the individual counting rate for a single detector—to be proportional to $\langle a^+a\rangle$. Therefore Eq. (6.14) yields the following expression for the ratio of the coincidence counting rate and the random coincidence counting rate, in the steady state:

$$\frac{\langle E^{(-)}(\mathbf{r},t)E^{(-)}(\mathbf{r},t+\tau)E^{(+)}(\mathbf{r},t+\tau)E^{(+)}(\mathbf{r},t)\rangle_{\rm st}}{\langle E^{(-)}(\mathbf{r},t)E^{(+)}(\mathbf{r},t)\rangle_{\rm st}^2} = \frac{\alpha_1(\tau)}{\alpha_1(\infty)}\frac{\rho_{22}(0\,|\,\tau)}{\rho_{22}(\infty)}\,, \tag{6.15}$$

where $\rho_{22}(0\,|\,\tau)[\equiv\alpha_1(\tau)]$ is the conditional probability mentioned above and where use has been made of Eq. (6.11).

Equation (6.15) tells us that the τ dependence of the coincidence counting rate equals that of $\rho_{22}(0\,|\,\tau)$. The latter quantity is readily derived from Eqs. (6.4) and (6.5) to be [Carmichael and Walls (1976a,b); cf. also Loudon (1980)]

$$\rho_{22}(0\,|\,\tau) = \rho_{22}(\infty)\left[1-\frac{3\gamma+\lambda}{2\lambda}\exp\left[-\frac{3\gamma-\lambda}{4}\tau\right] + \frac{3\gamma-\lambda}{2\lambda}\exp\left[-\frac{3\gamma+\lambda}{4}\tau\right]\right], \tag{6.16}$$

where the abbreviation

$$\lambda = (\gamma^2-16\Omega^2)^{1/2} \tag{6.17}$$

has been introduced, and

$$\Omega = 2\,|\,g\alpha\,| \tag{6.18}$$

denotes the Rabi frequency.

Of interest are two limiting cases of formula (6.16):

(a) Weak driving field ($\Omega \ll \gamma$). Then Eq. (6.16) takes the simple form

$$\frac{\rho_{22}(0\,|\,\tau)}{\rho_{22}(\infty)} = [1-\exp(-\tfrac{1}{2}\gamma\tau)]^2\ . \tag{6.19}$$

(b) Strong driving field ($\Omega \gg \gamma$). In this case Eq. (6.16) reduces to

$$\frac{\rho_{22}(0\,|\,\tau)}{\rho_{22}(\infty)} = 1-\cos\Omega t\,\exp(-\tfrac{3}{4}\gamma\tau)\ . \tag{6.20}$$

It becomes evident from Eq. (6.19) that the correlation time for the antibunching effect is roughly given, in the weak field case, by $2/\gamma$, i.e., twice the mean lifetime of the upper level with respect to spontaneous decay. When the driving field is very intense, one expects the "recovery time" for the atom to become shorter. In fact, this feature is expressed by Eq. (6.20), which indicates that $\rho_{22}(0\,|\,\tau)$ rises monotonically from zero to about half its maximum value during a time approximately given by $\tau=\pi/(2\Omega)$.

We thus have seen that resonance fluorescence radiation emitted by a single atom clearly exhibits the antibunching effect. This important feature is preserved, when the above assumption of an exactly resonant monochromatic exciting field is dropped, i.e., when allowance is made for detuning (Kimble and Mandel, 1976) and for the presence of a finite bandwidth associated with the driving field (Kimble and Mandel, 1977; Agarwal, 1978).

B. The effect of atomic number fluctuations

It has been shown in the preceding section that resonance fluorescence radiation from a single atom distinctly displays the antibunching phenomenon. Crucial to the generation of antibunching properties is the presence of only one atom. (To put it in experimental terms, care has to be taken that only light scattered by a single atom is focused on the detectors.) In fact, complete absence of nondelayed coincidence counts, as predicted in Sec. VI.A for the single-atom case, will no longer be observed when two or more atoms happen to be in the field of view, since two atoms will emit, with nonzero probability, a photon each at the same instant. Actually, in a recent experiment [Kimble, Dagenais, and Mandel (1977); Dagenais and Mandel (1978); a short description will be given in Sec. VII.B] the number of atoms in the observation volume could not be ensured to always be unity; instead it was subjected to fluctuations.

Hence there is considerable practical interest in the photon statistical properties of resonance radiation emitted by several atoms. The first to treat this problem were Agarwal *et al.* (1977), who analyzed collective atomic effects in resonance fluorescence from a few two-level atoms contained in a small volume. Theoretical studies closely connected with the experiment by Kimble, Dagenais, and Mandel (1977) are due to Jakeman *et al.* (1977), Carmichael *et al.* (1978), and Kimble, Dagenais, and Mandel (1978). Following the reasoning of the last group of authors (disregarding, however, the presence of background radiation that has been taken into account in all three references), we start from the observation that the delayed coincidence counting rate measured by means of two detectors with sufficiently small cathode areas in a Brown-and-Twiss—type arrangement equals, apart from a factor, the second-order correlation function

$$G^{(2)}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)=\langle E^{(-)}(\mathbf{r}_{\rm I},t)E^{(-)}(\mathbf{r}_{\rm II},t+\tau)\times E^{(+)}(\mathbf{r}_{\rm II},t+\tau)E^{(+)}(\mathbf{r}_{\rm I},t)\rangle\ , \tag{6.21}$$

where $\mathbf{r}_{\rm I}$ and $\mathbf{r}_{\rm II}$ are the detector positions.

Let us first assume the number of atoms M in the field of view to be fixed. The electric field at $\mathbf{r}(=\mathbf{r}_{\rm I},\mathbf{r}_{\rm II})$ is the sum of contributions due to the individual atoms

$$E^{(\pm)}(\mathbf{r},t)=\sum_{k=1}^{M}E_k^{(\pm)}(\mathbf{r},t)\ , \tag{6.22}$$

where the subscript k labels the atoms. Since we suppose the latter to be localized at random positions that are separated by many wavelengths in general, the operators $E_k^{(\pm)}$ involve phase factors with random phases, due to the geometrical factor $f(\mathbf{r})$ in Eqs. (6.1). Hence nonvanishing contributions to $G^{(2)}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)$ come only from pairings of the fields associated with the same atom. Thus insertion of Eq. (6.22) into Eq. (6.21) yields

$$\begin{aligned}G^{(2)}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)=&\sum_k\langle E_k^{(-)}(\mathbf{r}_{\rm I},t)E_k^{(-)}(\mathbf{r}_{\rm II},t+\tau)E_k^{(+)}(\mathbf{r}_{\rm II},t+\tau)E_k^{(+)}(\mathbf{r}_{\rm I},t)\rangle\\&+\sum_{k\neq l}\langle E_k^{(-)}(\mathbf{r}_{\rm I},t)E_k^{(+)}(\mathbf{r}_{\rm I},t)\rangle\langle E_l^{(-)}(\mathbf{r}_{\rm II},t+\tau)E_l^{(+)}(\mathbf{r}_{\rm II},t+\tau)\rangle\\&+\sum_{k\neq l}\langle E_k^{(-)}(\mathbf{r}_{\rm I},t)E_k^{(+)}(\mathbf{r}_{\rm II},t+\tau)\rangle\langle E_l^{(-)}(\mathbf{r}_{\rm II},t+\tau)E_l^{(+)}(\mathbf{r}_{\rm I},t)\rangle\ .\end{aligned} \tag{6.23}$$

Here we have assumed that the emission processes at two atoms are uncorrelated, which has allowed us to factorize the terms in the double sums.

When the detectors are at different positions $\mathbf{r}_{\rm I}\neq\mathbf{r}_{\rm II}$ (experimentally, this means that the two detectors are not perfectly aligned in conjugate position in a Brown-and-Twiss—type arrangement), the expectation values $\langle E_k^{(-)}(\mathbf{r}_{\rm I},t)E_k^{(+)}(\mathbf{r}_{\rm II},t+\tau)\rangle$ involve phase factors that vary with the position of the kth atom. As a consequence, the terms in the last sum in Eq. (6.23) will vanish on the statistical average. The terms in both the first and the second sum, on the other hand, are virtually independent of k (or l) and the detector positions, since the geometrical phase factors cancel. Hence, specializing to steady-state conditions, we arrive at the result

$$G_M^{(2)}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)=MG_1^{(2)}(\mathbf{r},\mathbf{r};t,t+\tau)+M(M-1)I_1^2\ . \tag{6.24}$$

Here, the subscript (M or 1) indicates the number of atoms contributing to the field, and I denotes the (mean) intensity.

Atomic number fluctuations are now readily taken into account by averaging Eq. (6.24) over M. In the case of a Poisson distribution, $\overline{M(M-1)}=\overline{M}^2$, where the bar denotes the averaging process, we thus obtain

$$\overline{G_M^{(2)}}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)=\overline{M}G_1^{(2)}(\mathbf{r},\mathbf{r};t,t+\tau)+\overline{M}^2I_1^2\ . \tag{6.25}$$

Hence the relative coincidence counting rate reads

$$\frac{\overline{K}_M(\tau)}{\overline{R}'^2_M\tau_{\rm resp}}=\frac{\overline{G_M^{(2)}}(\mathbf{r}_{\rm I},\mathbf{r}_{\rm II};t,t+\tau)}{\overline{I}_M^2}=\frac{1}{\overline{M}}\frac{K_1(\tau)}{R'^2_1\tau_{\rm resp}}+1\ , \tag{6.26}$$

(K being the coincidence counting rate and R' the single counting rate). Here use has been made of the relation

$$\overline{I}_M=\overline{M}I_1\ , \tag{6.27}$$

which is valid for atoms located at random positions.

The result (6.26) makes clear that the antibunching effect exhibited by the field due to a single atom [$K_1(0)=0$ according to Eq. (6.3)] is completely washed out by the atomic number fluctuations, since the coincidence counting rate equals the random coincidence counting rate at $\tau=0$ and is even greater for $\tau>0$. It should be noticed that this holds true irrespective of the actual value of $\overline{M}$. Hence, in an experiment aimed to demonstrate photon antibunching, it does not suffice to keep $\overline{M}$ in the order of unity; it is the disturbance due to the fluctuations of M that prevents the observation of the antibunching effect, in the sense of the definition given in Sec. II.A.

It should be emphasized, however, that Eq. (6.26), anyway, describes a nonclassical photon statistical feature: since $K_1(\tau)$ has its absolute mininum at $\tau=0$, Eq. (6.26) predicts the rise of the coincidence counting rate when τ grows from zero also in the case of a fluctuating number of atoms. In fact, such a behavior contradicts the fundamental classical inequality (2.3). It appears natural that this effect becomes less pronounced when $\overline{M}$ increases, as it is indicated by Eq. (6.26). Hence in an actual experiment the requirement $\overline{M} \lesssim 1$ should be met.

VII. SOME EXPERIMENTAL ASPECTS

A. Feasibility of experiments utilizing a nonlinear interaction

Let us now look at the various interaction mechanisms studied theoretically in Secs. III–V with an experimentalist's eye. A natural question arises: which type of experiments aimed to provide evidence of the antibunching effect might really be feasible?

To begin with, we briefly discuss the suitability of two-photon absorption for that purpose. Here a severe obstacle is posed by the low-intensity requirement, which is dictated by the fact that photon antibunching is a $1/\bar{n}$ effect, where $\bar{n}$ denotes the mean number of photons contained in the mode volume [cf. Eq. (2.24)]. The dilemma is that, on the one hand, a high intensity is needed for the two-photon absorption process to take place with appreciable efficiency, and, on the other hand, one-photon absorption cannot be used for the required reduction of the two-photon absorbed field, since the relative excess coincidence counting rate remains invariant under this type of attenuation, as has been outlined in Sec. III.A.

It would be desirable to produce the low-intensity field displaying antibunching by the two-photon absorption process itself, but the corresponding cross sections in known materials are far too small to allow for such a possibility, as will be seen from the following simple estimate.

When the mode volume V of cross-sectional area f and length $c\tau_{\rm crit}$ (see Sec. III.F) is initially filled with n photons, the photon flux per unit area j equals

$$j=\frac{n}{f\tau_{\rm crit}} . \tag{7.1}$$

As is well known, the probability, per unit time, w that an atom will undergo two-photon absorption can be written as

$$w=\sigma j^2 . \tag{7.2}$$

Hence, during its passage through the absorption cell of length L, the field contained in the mode volume will excite about

$$Z=w\frac{L}{c}N_{\rm at} \tag{7.3}$$

atoms, where $N_{\rm at}$ denotes the number of atoms per mode volume

$$N_{\rm at}=n_{\rm at}V , \tag{7.4}$$

$n_{\rm at}$ being the density of atoms. [Note that formula (7.3) actually overestimates Z when the photon number becomes considerably reduced in the course of interaction.]

From Eqs. (7.1)–(7.4) we find the number of photons δn that are absorbed from the mode volume while the field passes through the absorbing medium, to be

$$\delta n=2Z=2\sigma\frac{n^2}{f^2\tau_{\rm crit}^2}\frac{L}{c}n_{\rm at}V . \tag{7.5}$$

With the specification of the transverse dimension of the mode volume as given by Eq. (3.49), we may put

$$f\approx 2L\lambda, \quad V\approx 2L\lambda c\tau_{\rm crit} . \tag{7.6}$$

Hence Eq. (7.5) can be rewritten as

$$\delta n=\sigma\frac{n^2}{\lambda\tau_{\rm crit}}n_{\rm at} . \tag{7.7}$$

Since a considerable attentuation is needed to generate antibunching properties, we require the absorber to reduce the photon number n from, say, 10^3 to 10^2. Equation (7.7) thus gives us the following lower bound for the two-photon absorption cross section:

$$\sigma\gtrsim\frac{\lambda\tau_{\rm crit}}{10^3 n_{\rm at}} . \tag{7.8}$$

Putting here $\lambda=5\times10^{-5}$ cm, $\tau_{\rm crit}\approx10^{-9}$ s, and $n_{\rm at}\approx10^{20}$ cm^{-3}, we find this bound to be about 5×10^{-37} cm^4 s. However, this figure is greater by many orders of magnitude than that offered by nature. For rhodamine 6G, for example, σ takes the value 3×10^{-48} cm^4 s [see Hermann and Ducuing (1972)], the discrepancy thus exceeding 10 orders of magnitude!

By the way, it should be noticed that the situation is not improved when the absorption cell is made longer. [Note that the cell length L has actually disappeared from Eq. (7.8)]. This is due to the fact that an increase in the cell length gives rise to a similar growth of the cross-sectional area of the mode volume, as has been pointed out in Sec. III.F.

Insertion of the two-photon absorber into a cavity which either contains an amplifying (laser) medium (Bandilla and Ritze, 1976b) or is pumped by an external driving field (Chaturvedi, Drummond, and Walls, 1977) provides also no practical means of demonstrating the antibunching phenomenon. This technique severely suffers from the drawback that the process of coupling out the field, being required for an actual observation, has the character of an attenuation due to one-photon absorption, and hence leaves invariant the relative excess coincidence counting rate (see Sec. III.A), which is rather small because of the high number of photons present in the cavity.

A possible way out of the dilemma might be provided, at least in principle, by utilizing the technique of destructive interference for further attenuation of the two-photon absorbed light. In fact, it has been shown in Sec. V.B that the relative magnitude of the antibunching effect, as expressed by the quantity $\Delta/\bar{n}\equiv(\Delta n^2-\bar{n})/\bar{n}$, is essentially preserved in this process, under suitably chosen experimental conditions. However, one cannot expect that an attenuation by so many orders of magnitude as are needed in case of two-photon absorption could be accomplished by means of any practical interference device. The obstacles are inevitable imperfections of the optical elements (for instance, the maximum and the minimum transmittances of a polarizer differ from their ideal values 1 and 0, respectively) and limited mechanical stability of the setup. Hence the extreme precision needed in the proper adjustment of amplitudes and (relative) phases in order to attain drastic attenuation (by many orders of magnitude), due to interference, cannot be achieved in practice. Moreover, in the theoretical analysis we have idealized the light beams as monochromatic plane waves, thus neglecting the finite beam diameter, the angular spread in the direction of propagation, and the variation of the amplitude over the cross-sectional area. A different problem is the generation of strictly coherent light beams (being in Glauber states), since laser light exhibits amplitude fluctuations, in addition to phase diffusion, which are small well above threshold but nevertheless nonvanishing.

For all these reasons we can rule out two-photon (and with still more justification three-photon, etc.) absorption as a practical means of demonstrating the antibunching effect.

Similarly, techniques that make use of the Kerr effect are lacking practical relevance, since they also require drastic attenuation by destructive interference. This becomes evident from the analysis in Sec. V.C, where account is taken of the very weak coupling of the field to the Kerr medium. In fact, according to Eimerl (1978), a typical value for the coefficient $\Delta\omega$ in Eq. (5.17) is $\Delta\omega=\frac{1}{2}10^{-2}\ \mathrm{s}^{-1}$ (for liquid CS_2, the mode volume being taken as 1 mm^3).

With this figure, the quantity ε defined by Eq. (5.35), is about 10^{-12} for a cell length of 6 cm. It then follows from Eqs. (5.39) that the mean initial photon number A^2 should be of the order of 10^{12}, and this value should be reduced, after the beam has passed through the Kerr cell, to only a few photons, by destructive interference with a coherent reference beam, in order to obtain a field with antibunching properties. Actually, this procedure is not feasible, since according to Eqs. (5.42) it requires extremely small values (on the order of 10^{-6}) for both the relative phase and the deviation of the amplitude ratio for the two interfering beams from 1 to be set.

In contrast to the physical mechanisms considered thus far, parametric three-wave interaction has the advantage that the field that will acquire antibunching properties might be chosen as weak, as it will be needed for an actual observation of the antibunching phenomenon. Formally, this is due to the fact that the equations of motion, in the approximation of a prescribed (very intense) harmonic or sum-frequency wave, are linear in the photon creation and annihilation operators, respectively, for the waves of interest [see Eqs. (4.1), (4.2) and (4.14), (4.15)]. As a consequence, the relevant physical quantities $\Delta(t)/\bar{n}(t)$ (in the degenerate case) and $\Delta_{\mathrm{cross}}(t)/\bar{n}_s(t)$ (in the nondegenerate case) are practically independent of the mean photon number in the initial field(s), provided this figure is high enough to make the contributions originating from parametric fluorescence negligibly small. This has been shown in some detail in Secs. IV.A and B. Physically, the essential point is that in the presence of an intense pump wave (second harmonic or sum-frequency field) the effective cross section for the process leading to antibunching properties is drastically enhanced. In fact, the coupling constant γ in Eqs. (4.1), (4.2) or (4.14), (4.15) is proportional to the amplitude of the pump field E_p. For an estimate we may write [see, for example, Brunner and Paul (1977)]

$$\gamma\approx 4\pi\chi\omega E_p/\mu^2\ , \tag{7.9}$$

where χ is the nonlinear susceptibility, ω the circular frequency in the pump wave, and μ the linear index of refraction.

Expressing E_p through the energy flux per unit area ϕ_p,

$$E_p=\left[\frac{2\pi\phi_p}{\mu c}\right]^{1/2}, \tag{7.10}$$

we can replace Eq. (7.9) by

$$\gamma\approx 2(2\pi)^{3/2}\mu^{-5/2}c^{-1/2}\chi\omega\phi_p^{1/2}\ . \tag{7.11}$$

The numerical results presented in Sec. IV.A (see Table I) indicate that favorable conditions for the generation of antibunching properties correspond to an interaction time t_i that satisfies the condition $\gamma t_i\approx 0.4$. Hence, for a crystal length of about 4 cm, γ is required to be approximately $3\times 10^9\ \mathrm{s}^{-1}$. Thus Eq. (7.11) gives us the following estimate for ϕ_p

$$\phi_p\approx\frac{\mu^5 c}{4(2\pi)^3\omega^2\chi^2}10^{19}\ (\text{in esu})\ . \tag{7.12}$$

Inserting here a realistic value for the nonlinear susceptibility, say, $\chi\approx 10^{-8}$ esu [see for instance, Zernike and Midwinter (1973)], we find, for $\omega\approx 4\times 10^{15}\ \mathrm{s}^{-1}$ and $\mu\approx 1.5$,

$$\phi_p\approx 10^{12}\ \mathrm{erg\,cm^{-2}\,s^{-1}}=10^5\ \mathrm{W\,cm^{-2}}\ . \tag{7.13}$$

[A similar result has been obtained by Stoler (1974).]

Actually, such a figure can be attained by means of existing laser technology without too much effort. A specific difficulty, however, arises from the extremely short correlation time τ_{crit} for the parametric interaction process, which has been estimated in Sec. IV.C to be on the order of picoseconds or even shorter. In fact, an observation of the antibunching phenomenon becomes feasible only when available detectors' response time does not exceed τ_{crit}, since otherwise the effect under study will be wiped out. However, detectors with a response time as short as a few picoseconds do not exist. Nevertheless, there is a way of overcoming this difficulty afforded by modern laser technology which makes possible the generation of picosecond pulses in a cw regime. In those circumstances the detectors are only required to have a response time that is shorter than the time interval Δt between two successive pulses. This condition being fulfilled, they actually integrate over one pulse only. Since Δt is typically of the order of nanoseconds, the aforementioned requirement is easily met.

Specializing in the nondegenerate parametric process which offers better prospects than the degenerate one, owing to its longer correlation time, one might proceed as follows [improved version, due to Chmela, Horák, and Peřina (1981),[15] of an experiment proposed by Paul and Brunner (1980)]: two beams originating from two synchronously pumped cw picosecond lasers emitting at ω_s and ω_i, respectively, are made to generate an intense sum-frequency (pump) wave in a nonlinear crystal (see Fig. 9). The latter wave passes through a second nonlinear crystal similar to the first one, together with a signal and an idler wave that have been separated with the help of a beam splitter from the original laser beams and afterwards attenuated by means of a one-photon absorber so that each single pulse, in both trains, contains, say, about 2.5 photons on average, when entering the nonlinear crystal. Of course, the three pulse trains have to be synchronized. Moreover, their phases have to be adjusted in such a way that maximum amplification of the pump wave results. The experimental parameters should be chosen such that the condition $\gamma t_i = 0.35$ will be satisfied, ensuring that the antibunching effect displayed by the combined signal and idler output field becomes as large as possible for the initial photon numbers $\bar{n}_s(0) = \bar{n}_i(0) = 2.5$ (see Sec. IV.B). The pulse duration should be adjusted to the length of the second crystal such that it equals the correlation time given by Eq. (4.28).

To provide experimental evidence of photon antibunching, two techniques might be adopted. The first would be to count the photons contained in both a signal and a corresponding idler pulse. Then the antibunching effect would show up in a distribution for the number of photons thus registered that is narrower than a Poisson distribution. The second method would utilize the Brown-and-Twiss coincidence counting technique. The coincidence counting rate should be determined both for zero delay time and for a delay time corresponding to the distance between two different pulses, the antibunching effect being demonstrated by an excess of delayed coincidences.

It should be emphasized, however, that the feasibility of the proposed demonstration of photon antibunching critically relies upon the assumption that cw picosecond pulse trains can be generated which are ideal in the sense that the pulses are precise replica of each other. Formally, this means that a generalized Glauber state—i.e., a Glauber state with respect to a nonmonochromatic mode [cf. Titulaer and Glauber (1966)]— can be ascribed to the ensemble constituted by all the single pulses in the train. Experimentally, the basic requirement is that the photon statistics of the original cw picosecond pulse trains, to be measured after sufficiently strong attenuation through one-photon absorption using one of the above-mentioned techniques, have Poissonian character to a very good approximation.

Moreover, the requirement of preventing pump photons from reaching the detectors, i.e., of filtering out the pump wave, poses a serious problem to the experimentalist, too, since the intensity of this wave exceeds that of the waves that are ultimately of interest, by many orders of magnitude.

Hence one should not be too optimistic with regard to the prospects for the experiment under discussion.

B. Measurement of intensity correlations in resonance fluorescence

Resonance fluorescence (from a single atom) differs favorably from the nonlinear interaction mechanisms discussed in the foregoing in that the light thus generated is already endowed with the desired antibunching properties, as has been pointed out in Sec. VI.A. Hence, from the experimental point of view, this process appears to be the most promising one. In fact, it has already been studied experimentally by Kimble, Dagenais, and Mandel (1977) and Dagenais and Mandel (1978). Moreover, the results of a similar experiment by Leuchs, Rateike, and Walther have recently been reported in a review article by Walls (1979). In the following, I will briefly describe the experiment of Mandel and his co-workers and discuss their findings.

In order to keep the average number of atoms in the field of view small (preferably below unity), the authors used an atomic beam with the following physical parameters: width 100 μm, mean velocity about 10^5 cm s^{-1}, and flux density 10^{10} to 10^{11} atoms per cm^2 and s. With the help of a microscope objective they collected the fluorescence radiation from a region whose linear dimension was about 100 μm, in a direction at right angles both to the atomic beam and to the laser beam driving the atoms. The collected light, after passing a beam

[15]Those authors have also suggested more sophisticated schemes for the measurement of antibunching in nonlinear optical processes.

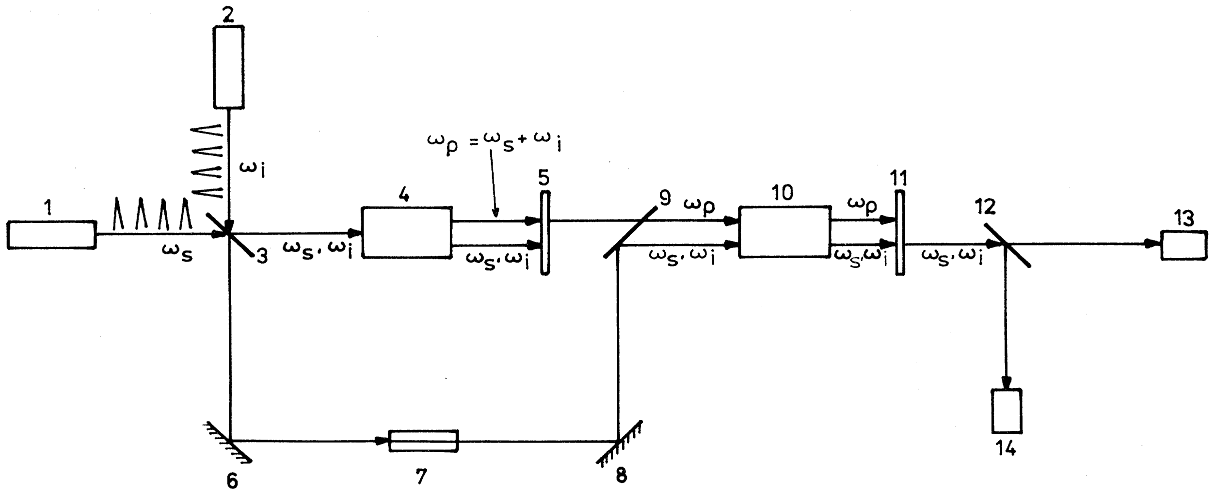

FIG. 9. Proposed setup for the observation of photon antibunching in parametric three-wave interaction. 1, 2, cw picosecond lasers emitting at ω_s and ω_i, respectively; 3, 9, 12, beam splitters; 4, 10, nonlinear crystals; 5, 11, filters; 6, 8, mirrors; 7, one-photon absorber; 13, 14, detectors.

splitter, was focused on the entrance apertures of two photon counters.

The atomic beam consisted of sodium atoms. Before being subjected to the laser field that excited resonance fluorescence, they were prepared in the $3^2S_{1/2}$, $F=2$, $m_F=2$ magnetic sublevel by means of optical prepumping, in a weak magnetic field, with a tunable dye laser, followed by spontaneous downward transitions. From this level only the transition to the $3^2P_{3/2}$, $F=3$, $m_F=3$ sublevel is allowed. Hence irradiating the atoms with a laser beam that is resonant to this transition (and accurately orthogonal to the atomic beam, in order to minimize Doppler shifts as "seen" from an atom), the authors ensured that the atoms behaved as two-level systems in the process of resonance fluorescence under observation.

From the above-mentioned figures for the atomic velocity and the dimensions of the observation region it follows that the transit time of an atom through this region is about 100 ns. Hence transit-time corrections, as they have been included in their analysis by Kimble, Dagenais, and Mandel (1978), become important as the delay time approaches 100 ns.

Photon counting has been performed by using the time-to-digital-converter (TDC) technique. (The pulses from the first detector are fed to the start input of the converter, whereas the second detector sends its pulses to the stop input.) The TDC effectively digitized the time intervals τ between start and stop pulses in units of 0.5 ns.

It should be noticed that the number of pulse pairs thus recorded in a given delay channel τ is not strictly a measure of the second-order correlation function $G^{(2)}(t,t+\tau)$. The point is that $s^2G^{(2)}(t,t+\tau)$, where s is the detector sensitivity, describes the joint probability, per (unit time)^2, for the detection of two photons at time t and $t+\tau$, respectively, irrespective of whether, in the meantime, counts are triggered on the detectors, whereas the operational mode of a TDC makes sure that no photon is recorded by the stop detector in the time interval between the two counts that are registered, with the help of the TDC technique, as a delayed coincidence. Hence the use of a TDC will lead to an underestimation of the true coincidence counting rate for large values of τ and/or high intensities. In order to estimate this error one has to compare the joint probability, per (unit time)^2, $s^2G^{(2)}(t,t+\tau)$ with the probability, per (unit time)^2, $W(A,B,C)$ that the following three events will occur: (a) detection of *one* photon at time t, (b) detection of *no* photon in the time interval t to $t+\tau$, (c) detection of *one* photon at time $t+\tau$.

An explicit expression for $W(A,B,C)$ has been derived in case of a classical field (Troup, 1966), or, a little more generally, a quantum field that allows for a P representation (Barakat and Blake, 1980). One easily infers from those formulas that the deviation of $W(A,B,C)$ from $s^2G^{(2)}(t,t+\tau)$ is negligibly small when the condition

$$s\int_t^{t+\tau}\langle E^{(-)}(t')E^{(+)}(t')\rangle dt' \ll 1 \tag{7.14}$$

is fulfilled, which evidently means that the average number of photon counts during the interval t to $t+\tau$ should be small in comparison to unity.

Fortunately, in the experiment under consideration the mean counting rates of the two photodetectors were low enough (about 2×10^4 s^{-1}) to ensure the validity of the inequality (7.14) even for values of τ as large as 100 ns. Hence it is justified, under those specific conditions, to take the number of pulse pairs registered in the delay channel τ as a measure of the second-order correlation function $G^{(2)}(t,t+\tau)$, as has actually been done by the authors who performed the experiment.

Their experimental results are represented in Fig. 10. Here the number of registered pulse pairs $n(\tau)$ is plotted

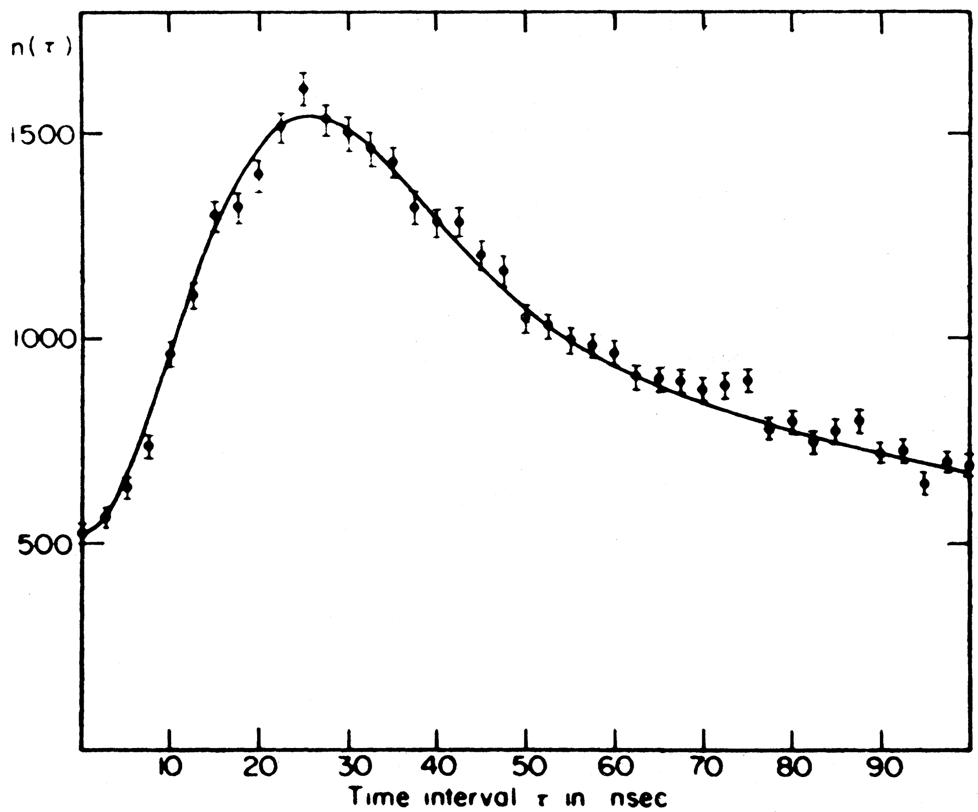

FIG. 10. Measured number of photon coincidences $n(\tau)$ vs delay time. The curve represents the theoretical predictions. [After Dagenais and Mandel (1978).]

versus the delay time τ. The characteristic feature displayed by this curve is the pronounced positive slope near $\tau=0$. As already mentioned in Sec. VI.B, this behavior is nonclassical, since it is in contradiction to the inequality (2.3) quite generally valid in classical wave theory. The value of $n(0)$ is greater, however, than that characteristic of random coincidences. As has been pointed out in Sec. VI.B, this result is easily understood as a consequence of atomic number fluctuations which, in fact, cannot be excluded when an atomic beam technique is used.

Dagenais and Mandel (1978) inferred from their experimental parameters that the average number of atoms in the field of view was as low as 0.37. Moreover, they extracted from their data, by comparison with their theoretical predictions (Kimble, Dagenais, and Mandel, 1978), the second-order correlation function for the resonance radiation due to precisely one atom. They thus obtained the values displayed in Fig. 11, which fit very well to the theoretical curves.

Let us now turn to the somewhat delicate question of whether convincing evidence of photon antibunching has been provided in the experiment under consideration. There is, of course, no doubt that this effect was observed at least indirectly, as indicated by Fig. 11. Adhering, however, as most workers in this field do, to the definition of antibunching as a deficit of (nondelayed) coincidences in comparison to random coincidences, one cannot take the above-mentioned experimental results as a direct demonstration of the antibunching phenomenon, since the measured excess coincidence counting rate was actually positive, as it must be, in the presence of atomic number fluctuations, according to the theoretical analysis given in Sec. VI.B. Only if one is willing to interpret the mere occurrence of a dip at $\tau=0$ in the curve displaying the number of coincidence counts registered (see Fig. 10) as an indication of photon antibunching (which, in fact, one is tempted to do when visualizing the literal meaning of the term "antibunching"), will one agree with Kimble, Dagenais, and Mandel (1977), who claimed to have provided "unmistakable evidence" for photon antibunching. Anyway, however, their experimental findings, contradicting, as they do, the classical inequality (2.3), reflect a specific nonclassical feature of the radiation field.

C. Time correlations and spectral features

There is a well-known complementarity between time and frequency measurements: observing, with the help of a photodetector, the emission times of photons (strictly speaking, the times of arrival on the detector), one deprives oneself, on principle, of gaining information on

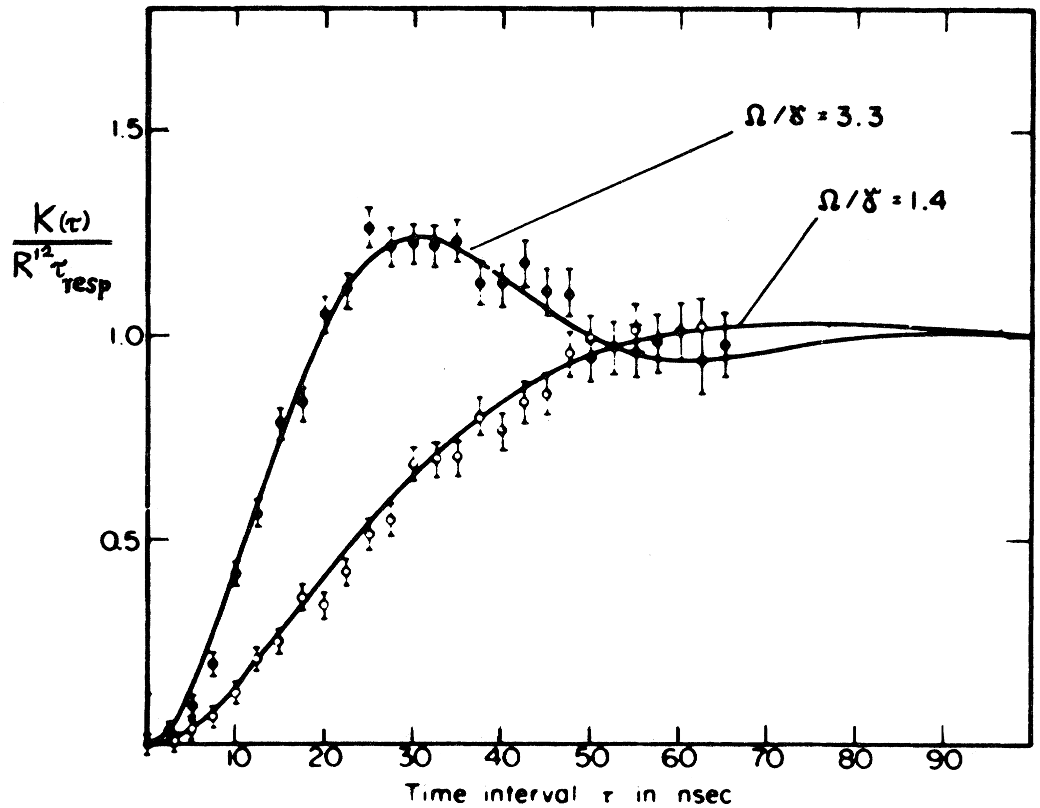

FIG. 11. The relative coincidence counting rate (with respect to the random coincidences) $K(\tau)/(R'^2\tau_{\text{resp}})$ for a single atom, as derived from the measurements, for two different values for the ratio of the Rabi frequency Ω and the spontaneous radiative decay rate of the upper level γ. [After Dagenais and Mandel (1978).]

the spectrum of those photons. On the other hand, the accuracy in determining the spectral features of the emitted light is bought with the loss of information on the emission time of the photons that are registered in the spectrometer. However, when lowering the precision requirements on both the spectral and the temporal characteristics, a compromise has been found in the case of resonance fluorescence which allows the analysis of time correlations between photons belonging to different sidebands in the emission spectrum. Although this problem is only loosely connected with photon antibunching, I shall include a brief description of the experiment in question (Aspect *et al.*, 1980), in view of its surprising result and the general interest it deserves.

A dozen years ago, Mollow (1969) predicted that the resonance fluorescence spectrum of a strongly driven two-level atom would consist of three components, i.e., that it would constitute a triplet. Meanwhile, this phenomenon has been observed by several authors, the first to do so being Schuda, Stroud, and Hercher (1974). The idea of the experiment under discussion [Cohen-Tannoudji and Reynaud (1979); see also Apanasevich and Kilin (1979)] is to equip the detectors in a coincidence counting device with filters which select photons from the higher-frequency and the lower-frequency sideband, respectively. The experimental conditions were chosen such that the frequency resolution of the filters $\Delta\nu$ obeyed the inequalities $\gamma \ll \Delta\nu \ll \Omega$, where γ is the width of the sidebands and Ω their separation. In order to make Ω sufficiently large, the laser frequency ω_L was detuned from the atomic frequency ω_0 towards higher frequencies by an amount much greater than the Rabi notation frequency. In those circumstances, the emission spectrum exhibited a pronounced peak at ω_L (Rayleigh scattering) and two lower-intensity sidebands centered at $2\omega_L - \omega_0$ and ω_0, respectively. The widths of the sidebands γ are given by that of the upper atomic level.

In accordance with theoretical predictions (Cohen-Tannoudji and Reynaud, 1979; Apanasevich and Kilin, 1979) the outcome of the experiment gave evidence of a strong temporal correlation between the frequency-selected photons, indicating that an atom, having emitted a photon into the higher-frequency sideband, emits the next photon into the lower-frequency sideband.

This interesting result is easily explained in a perturbative approach (see Aspect *et al.*, 1980): the occurrence of the two sidebands, in the off-resonance case studied experimentally, is due to a four-photon process with an intermediate resonance at the upper atomic level. In a first step, two laser photons are absorbed, and one photon corresponding to the higher-frequency sideband is emitted, the atomic system thus being brought into the upper level; and in a second step the atom spontaneously emits a photon corresponding to the lower-frequency sideband. It is interesting to note that in the present case the registration of the higher-frequency photon witnesses of the occurrence of a jump from the lower to the upper atomic level through a three-photon process, while, on the contrary, in the absence of filters (antibunching experiment) the registration of a photon is always associated with a transition from the upper to the lower level.

Theoretical studies (Cohen-Tannoudji and Reynaud, 1979; Apanasevich and Kilin, 1979) lead to the prediction that in the resonance case, at high intensities of the driving field, the above-mentioned temporal order of the emitted photons disappears. In any case, however, the correlations between photons emitted into different sidebands are of bunching type (the coincidence counting rate is maximum at zero delay time), while photons emitted into the same sideband will exhibit the antibunching effect (Apanasevich and Kilin, 1979).

D. Concluding remarks

It has been pointed out in Sec. VII.B that the actual uncertainty, unavoidable in an atomic beam experiment, of the number of atoms in the field of view prevents a direct observation of photon antibunching, in the sense that the (nondelayed) coincidence counting rate will be found to be lower than the accidental one. However, in the last few years techniques have been developed that may allow researchers to overcome this specific difficulty. In fact, recently Neuhauser *et al.* (1980) succeeded in confining a single barium ion in a rf quadrupole trap with a lifetime that could be made unlimitedly large by means of optical sideband cooling. Even without cooling, the ion could be held for times as long as 30 s. Studying the resonance fluorescence radiation from such a localized ion then should make possible a convincing demonstration of the antibunching effect.

Finally, I should like to emphasize that nonclassical behavior of light in the form of two beams has already been observed in earlier experiments. Specifically, it has been shown experimentally that the classical inequality

$$(\overline{I_1 I_2})^2 \leq \overline{I_1^2 I_2^2} \tag{7.15}$$

(I_1 and I_2 being the instantaneous intensities in the two beams), which is similar[16] to (4.24), is violated in certain circumstances when the cross and autocorrelation functions are measured with the help of the coincidence counting technique. [For more details see the review article by Loudon (1980).] In fact, observing double-beam coincidence counts in two-photon cascade emission (the two beams being generated in successive atomic transitions, respectively, and hence differing in their frequencies), Clauser (1974) arrived at the result that the left-hand side of (7.15) was larger by a factor of about 5 at maximum than the right-hand side. A really drastic violation of (7.15) has been reported by Burnham and Weinberg (1970), who studied parametric fluorescence under such experimental conditions that only one elementary process producing both a single and an idler photon took place, on average, during a detection period. They found the left-hand side of (7.15) to exceed the right-hand side by a factor of 10^4!

Physically, it is clear that the reason for those discrepancies lies in the corpuscular nature of light emitted in definite single events. Hence the above-mentioned experiments add, as the demonstration of photon antibunching will do, to the observations that confirm the photon concept so ingeniously contrived by Einstein (1905).

[16]Like (4.24), the inequality (7.15) is easily proved with the help of Schwarz's inequality. Moreover, it is readily seen that (7.15) is implied by (4.24).

ACKNOWLEDGMENTS

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