Multiatom and transit-time effects on photon-correlation measurements in resonance fluorescence

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(Received 3 February 1978)

An expression is derived for the expected number of photon pairs separated by a time interval τ that are detected in photoelectric correlation measurements of an atomic beam, when due account is taken of the fluctuations of the number of radiating atoms and of the effect of their finite transit time through the field of view. The theoretical expression is checked against some recent measurements and good agreement is obtained.

I. INTRODUCTION

In a recent photoelectric-correlation experiment,[1] it was demonstrated that photons emitted by resonantly excited atoms in an atomic beam exhibit anticorrelation or antibunching, as predicted by quantum electrodynamics.[2,3] Although the results of the measurements were in qualitative agreement with the theory, quantitative agreement requires a generalization of the treatment to include the effect of a variable number of atoms and geometric corrections for the finite transit time through the field viewed by the photodetectors. The first effect becomes particularly important in correlation experiments at larger atomic fluxes, such as those reported by Walther,[4] but it plays a role even at very low beam currents. The transit-time corrections depend on the observed time intervals between photoelectric pulse pairs, and increase with increasing time delay. Finally, the corrections for multiatom effects are expected to differ according to whether a single detector[4] or a pair of photodetectors[1] is used in the experiment. Some discussion of the correction for a variable number of atoms has also been recently given by Jakeman *et al.*[5]

In the following, we generalize the treatment of photoelectric-correlation measurements of resonance fluorescence from an atomic beam to include these effects, and we show that good quantitative agreement between experiment and theory can be obtained.

II. CORRECTION FOR MULTIATOM CONTRIBUTIONS

In the experiments we are discussing, photons emitted by one or more atoms of an atomic beam, that are excited by a perpendicular, resonant beam of light from a tunable dye laser, are collected in a direction at right angles to both beams. They are directed by an optical system to fall on either two or one photodetector(s), depending on whether or not a beam splitter is used. The time interval τ between one photoelectric pulse occurring at time t, the "start" pulse a, and another pulse occurring at time $t+\tau$, the "stop" pulse b, is digitized with time resolution $\Delta\tau$, and the number of events $n(\tau)$ corresponding to pulse pairs with time separation τ is recorded over some long period. If $\mathfrak{N}$ is the number of accepted start pulses in the course of an experiment, and $\mathcal{P}(\tau)\Delta\tau$ is the conditional probability that a start pulse at time t is followed by a stop pulse at time $t+\tau$ within $\Delta\tau$, in the steady state, then the expectation of $n(\tau)$ is

$$\langle n(\tau)\rangle = \mathfrak{N}\Delta\tau\mathcal{P}(\tau). \tag{1}$$

From Glauber's treatment of the detection problem[6] it follows that $\mathcal{P}(\tau)$ is expressible in terms of the light intensity[7]

$$\hat{I}(\vec{\mathbf{r}},t) \equiv \hat{E}_i^{(-)}(\vec{\mathbf{r}},t)\cdot\hat{E}_i^{(+)}(\vec{\mathbf{r}},t)$$

at the photodetector(s), expressed in photons per unit area per second, by

$$\mathcal{P}(\tau) = \frac{\alpha_b\int_{S_a}\int_{S_b} d\vec{\mathbf{r}}_a\, d\vec{\mathbf{r}}_b \langle :\mathcal{T}\hat{I}(\vec{\mathbf{r}}_a,t)\hat{I}(\vec{\mathbf{r}}_b,t+\tau):\rangle}{\int_{S_a} d\vec{\mathbf{r}}_a\langle \hat{I}(\vec{\mathbf{r}}_a,t)\rangle}, \tag{2}$$

where $\mathcal{T}$ is the time-ordering operator and $\hat{E}_i^{(+)}(\vec{\mathbf{r}},t)$ is the positive frequency part of the electric field at $\vec{\mathbf{r}}$ at time t. Here α_a, α_b are dimensionless quantum efficiencies of the "start" and "stop" detectors, respectively, and the integrals are to be taken over the exposed surface areas S_a, S_b of the detectors, which may be different or one and the same. It is the presence of the normally ordered correlation function in Eq. (2) that accounts for the antibunching.

In practice, the field $\hat{E}_i(\vec{\mathbf{r}},t)$ at the detector con-

tains fluorescent contributions from several different atoms that may be in the field of view, and from the background or exciting light. Because the atomic separations are on the order of many wavelengths, cooperative atomic effects are expected to be very small and will be neglected. If N atoms contribute to the field, we therefore write

$$\hat{\vec{E}}_N^{(+)}(\vec{r},t)=\sum_{k=1}^{N}\hat{\vec{E}}^{(+)(k)}(\vec{r},t)+\vec{e}(\vec{r},t)\,, \tag{3}$$

where the sum is to be taken over all the atoms $k=1,2,\ldots,N$ that are in the field of view at time t (we ignore the transit time of the light), and we treat the background light $\vec{e}(\vec{r},t)$ from the laser as a c number. For the moment we regard N as a fixed number. The corresponding integrated mean light intensity is then given by

$$\int_{S_a}d\vec{r}_a\,\langle\hat{I}_N(\vec{r}_a,t)\rangle = \int_{S_a}d\vec{r}_a\Big\langle\Big(\sum_k\hat{E}_i^{(-)(k)}(\vec{r}_a,t)+e_i^*(\vec{r}_a,t)\Big)\times\Big(\sum_l\hat{E}_i^{(+)(l)}(\vec{r}_a,t)+e_i(\vec{r}_a,t)\Big)\Big\rangle\,, \tag{4}$$

with summation over repeated Cartesian indices i implied. For N atoms, Eq. (1) should be replaced by

$$\langle n_N(\tau)\rangle=\mathfrak{N}_N\Delta\tau\mathcal{P}_N(\tau). \tag{5}$$

Here $\mathfrak{N}_N$ is the number of start pulses with N atoms in the field of view, and $\mathcal{P}_N(\tau)$ is the corresponding conditional probability, which is given by an equation like Eq. (2) with suffix N on the light intensities. $\langle n_N(\tau)\rangle$ is the expected number of pulse pairs with time separation τ corresponding to N atoms.

Of the four terms contributing to the integral on the right of Eq. (4), we expect the two involving the correlations $\langle\hat{E}_i^{(-)(k)}e_i\rangle$ and $\langle e_i^*\hat{E}_i^{(+)(l)}\rangle$ to make a negligible contribution, as these correlations will be proportional to phase factors that oscillate rapidly with changing $\vec{r}_a$. Second, although we expect the fields radiated by different atoms to be partly correlated, we may reasonably ignore terms of the type $\langle\hat{E}_i^{(-)(k)}\hat{E}_i^{(+)(l)}\rangle$ with $k\neq l$ as small compared with the others. The reason is that the fluorescence is collected at right angles to the incident laser beam, and different atoms are located at random positions that are separated by many wavelengths in general, so that the cross-correlation is proportional to a rapidly oscillating phase factor that averages to zero. Finally, if we denote $\langle\hat{E}_i^{(-)(k)}(\vec{r}_a,t)\hat{E}_i^{(+)(k)}(\vec{r}_a,t)\rangle$ by $\langle\hat{I}_{1a}\rangle$ and $\langle e_i^*(\vec{r}_a,t)e_i(\vec{r}_a,t)\rangle$ by $\langle i_a\rangle$, and make the reasonable assumption that the first term is approximately the same for every atom in the field of view, and that neither term varies much with $\vec{r}_a$, then Eq. (4) reduces to

$$\int_{S_a}d\vec{r}_a\,\langle\hat{I}_N(\vec{r}_a,t)\rangle=S_a(N\langle\hat{I}_{1a}\rangle+\langle i_a\rangle)\,. \tag{6}$$

The intensity correlation function that enters in the definition of $\mathcal{P}_N(\tau)$ is more complicated. If we make use of Eq. (3) in the definition of the light intensity $\hat{I}_N(\vec{r},t)$, and then form the cross-correlation, we obtain 16 different terms. Of these, eight terms have unpaired $\hat{E}_i(\vec{r},t)$ or $e_i(\vec{r},t)$ factors and will be discarded, because we expect the corresponding surface integrals to make a negligible contribution by virtue of the rapidly oscillating phases. Two other terms of the form

$$\langle\hat{E}_i^{(-)(k)}(\vec{r}_a,t)\hat{E}_j^{(-)(l)}(\vec{r}_b,t+\tau)e_j(\vec{r}_b,t+\tau)e_i(\vec{r}_a,t)\rangle$$

and its complex conjugate, also have rapidly oscillating phases and will also be discarded. We are then left with the following expression:

$$\begin{aligned}\int_{S_a}\int_{S_b}d\vec{r}_a\,d\vec{r}_b\langle{:}\mathcal{T}\hat{I}_N(\vec{r}_a,t)\hat{I}_N(\vec{r}_b,t+\tau){:}\rangle &= \int_{S_a}\int_{S_b}d\vec{r}_a\,d\vec{r}_b\Big(\sum_k\sum_l\sum_m\sum_n\langle\hat{E}_i^{(-)(k)}(\vec{r}_a,t)\hat{E}_j^{(-)(l)}(\vec{r}_b,t+\tau)\hat{E}_j^{(+)(m)}(\vec{r}_b,t+\tau)\hat{E}_i^{(+)(n)}(\vec{r}_a,t)\rangle\\ &+\sum_k\sum_m\langle\hat{E}_i^{(-)(k)}(\vec{r}_a,t)\hat{E}_j^{(+)(m)}(\vec{r}_b,t+\tau)\rangle\langle e_j^*(\vec{r}_b,t+\tau)e_i(\vec{r}_a,t)\rangle+\text{c.c.}\\ &+\sum_k\sum_n\langle\hat{E}_i^{(-)(k)}(\vec{r}_a,t)\hat{E}_i^{(+)(n)}(\vec{r}_a,t)\rangle\langle e_j^*(\vec{r}_b,t+\tau)e_j(\vec{r}_b,t+\tau)\rangle\\ &+\sum_l\sum_m\langle\hat{E}_j^{(-)(l)}(\vec{r}_b,t+\tau)\hat{E}_j^{(+)(m)}(\vec{r}_b,t+\tau)\rangle\langle e_i^*(\vec{r}_a,t)e_i(\vec{r}_a,t)\rangle\\ &+\langle e_i^*(\vec{r}_a,t)e_j^*(\vec{r}_b,t+\tau)e_j(\vec{r}_b,t+\tau)e_i(\vec{r}_a,t)\rangle\Big)\,,\end{aligned} \tag{7}$$

where we have assumed that correlations involving the fluorescence and the background light may be factored. In Eq. (7), the sums over k and n, strictly speaking, are to be taken over the N atoms that are in the field of view at time t, and those over l and m over the atoms in the field of view at the later time $t+\tau$. These are not necessarily the same atoms.

If the fields produced by different atoms are assumed to have correlations that integrate almost to zero because of the random atomic positions, then the only nonvanishing contributions to Eq. (7) come from paired fields. Thus, the third term in Eq. (7)

$$= \int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b \sum_k \langle \hat{I}^{(k)}(\vec{r}_a, t)\rangle\langle i(\vec{r}_b, t+\tau)\rangle$$

$$= NS_aS_b\langle \hat{I}_{1a}\rangle\langle i_b\rangle , \tag{8}$$

and the fourth term of Eq. (7)

$$= \int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b \sum_l \langle \hat{I}^{(l)}(\vec{r}_b, t+\tau)\rangle\langle i(\vec{r}_a, t)\rangle$$

$$= NS_aS_b\langle \hat{I}_{1b}\rangle\langle i_a\rangle , \tag{9}$$

whereas the second term in Eq. (7)

$$= \int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b \left(\sum_k \langle \hat{E}_i^{(-)(k)}(\vec{r}_a, t)\hat{E}_j^{(+)(k)}(\vec{r}_b, t+\tau)\rangle\langle e_j^*(\vec{r}_b, t+\tau)e_i(\vec{r}_a, t)\rangle + \text{c.c.}\right), \tag{10}$$

which involves cross-correlation functions of the second order in both the fluorescent and the background light. Both correlations are expected to have rapidly oscillating phase factors that depend on positions $\vec{r}_a$, $\vec{r}_b$ and on τ. In the special case $\vec{r}_a = \vec{r}_b$, these phase factors cancel up to a constant, but this can happen only when we integrate over the same photodetector, and not when two detectors are used. In general, the first phase factor will vary with the position of the atom, and even with a single detector the contribution of this term is expected to be small and will be discarded.[8]

We now turn to the first term in Eq. (7). If the only nonvanishing contributions come from pairings of fields associated with the same atom, the first term in Eq. (7)

$$\begin{aligned} = \int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b \Bigg(&\sum_{k\neq l}\sum \langle \hat{E}_i^{(-)(k)}(\vec{r}_a, t)\hat{E}_j^{(+)(k)}(\vec{r}_b, t+\tau)\rangle\langle \hat{E}_j^{(-)(l)}(\vec{r}_b, t+\tau)\hat{E}_i^{(+)(l)}(\vec{r}_a, t)\rangle \\ &+ \sum_{k\neq l}\sum \langle \hat{E}_i^{(-)(k)}(\vec{r}_a, t)\rangle\langle \hat{E}_i^{(+)(k)}(\vec{r}_a, t)\rangle\langle \hat{E}_j^{(-)(l)}(\vec{r}_b, t+\tau)\hat{E}_j^{(+)(l)}(\vec{r}_b, t+\tau)\rangle \\ &+ \sum_k \langle \hat{E}_i^{(-)(k)}(\vec{r}_a, t)\hat{E}_j^{(-)(k)}(\vec{r}_b, t+\tau)E_j^{(+)(k)}(\vec{r}_b, t+\tau)E_i^{(+)(k)}(\vec{r}_a, t)\rangle \end{aligned}$$

$$\begin{aligned} = \int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b \Bigg(&\sum_{k\neq l}\sum \Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_b, \tau)\Gamma_{ij}^{(l)*}(\vec{r}_a, \vec{r}_b, \tau) + \sum_{k\neq l}\sum \langle \hat{I}^{(k)}(\vec{r}_a, t)\rangle\langle \hat{I}^{(l)}(\vec{r}_b, t+\tau)\rangle \\ &+ \sum_k \langle \hat{I}^{(k)}(\vec{r}_a, t)\rangle\langle \hat{I}^{(k)}(\vec{r}_b, t+\tau)\rangle[1+\lambda_1(\tau)]\Bigg), \end{aligned} \tag{11}$$

where we have neglected field commutators associated with fields from different atoms, $\Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_b, \tau)$ is the second-order correlation tensor of the field produced by the kth atom, and $\lambda_1(\tau)$ is the normalized intensity-correlation function of the fluorescence produced by one atom. The latter is independent of the position of the atom over the range of interest, because the corresponding phase factors cancel, and $\lambda_1(\tau)$ therefore does not need to carry the atomic label k. The form of $\lambda_1(\tau)$ was analyzed in detail in Ref. 3. Its most interesting feature is that it starts from the value -1 for $\tau = 0$, and this gives rise to the photon antibunching.

The product

$$\Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_b, \tau)\Gamma_{ij}^{(l)*}(\vec{r}_a, \vec{r}_b, \tau),$$

in general, involves a phase factor that varies with position, so long as $\vec{r}_a \neq \vec{r}_b$, and therefore makes a small contribution to the integral. On the other hand, when $\vec{r}_a = \vec{r}_b$ under the integral, which is possible when a single photodetector is used,

$$\Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_a, \tau) \approx \Gamma_{ij}^{(l)}(\vec{r}_a, \vec{r}_a, \tau)$$

and the product becomes real and reduces to

$$\Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_a, \tau)\Gamma_{ij}^{(l)*}(\vec{r}_a, \vec{r}_a, \tau) \approx \langle \hat{I}(\vec{r}_a)\rangle^2 |\gamma(\tau)|^2, \quad (12)$$

where $\gamma(\tau)$ is the normalized autocorrelation function of the fluorescent field. This is independent of atomic position and of $\vec{r}_a$ to a good approximation. More generally, the product in Eq. (11) is well approximated by $\langle \hat{I}(\vec{r}_a)\rangle^2 |\gamma(\tau)|^2$ when the points $\vec{r}_a$ and $\vec{r}_b$ lie within the same coherence area on the photocathode, and it makes a small contribution otherwise, so that we may write

$$\int_{S_a}\int_{S_b} d\vec{r}_a\, d\vec{r}_b\, \Gamma_{ij}^{(k)}(\vec{r}_a, \vec{r}_b, \tau)\Gamma_{ij}^{(l)*}(\vec{r}_a, \vec{r}_b, \tau) \approx \delta_{ab} S_a^2 \eta \langle \hat{I}_{1a}\rangle^2 |\gamma(\tau)|^2, \quad (13)$$

where $\eta \leq 1$ and $1/\eta$ is a measure of how many coherence areas fall within the cathode area S_a. For a sufficiently small detector area S_a, $\eta \approx 1$. δ_{ab} is unity when the two photocathodes coincide, i.e., when a single photodetector is used, and zero otherwise. We therefore have from Eqs. (11) and (13) for the first term in Eq. (7)

$$N\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle S_a S_b \times [\delta_{ab}\eta(N-1)|\gamma(\tau)|^2 + (N-1) + (1+\lambda_1(\tau))]. \quad (14)$$

Finally, for the fifth term in Eq. (7) we evidently have

$$S_a S_b \langle i_a\rangle \langle i_b\rangle. \quad (15)$$

When the results of Eqs. (8)–(10), (14), and (15) are combined with Eqs. (5) and (6) we arrive at

$$\langle n_N(\tau)\rangle = \mathfrak{N}_N \Delta\tau \alpha_b S_b \left(N\langle \hat{I}_{1b}\rangle + \langle i_b\rangle + \frac{N\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle [1+\lambda_1(\tau)] - N\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle + \delta_{ab}\eta N(N-1)\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle |\gamma(\tau)|^2}{N\langle \hat{I}_{1a}\rangle + \langle i_a\rangle} \right) \quad (16)$$

for the expected number of events corresponding to a time interval τ when there are N atoms in the field of view.

It is worth noting that when N is very large, and when a single very small area photodetector is used, so that $\delta_{ab}\eta \approx 1$, then the term with $|\gamma(\tau)|^2$ is dominant in the numerator in Eq. (16), and the resulting expression reduces to the familiar form for light obeying thermal statistics.[9] In this many-atom limit the photons will exhibit bunching rather than antibunching, as was already suggested by Carmichael and Walls.[2]

III. TRANSIT-TIME CORRECTIONS

In practice, the fluorescent light is radiated by an atom moving with some velocity v across the field of view of finite transverse length l, so that the fluorescence from one atom can only be detected over a limited time. This effect reduces the contributions of single atoms to the measured correlation function in Eq. (16). If two photons with time separation τ from one atom are to be detected, the atom has to radiate the first photon in the initial distance $l - v\tau$ along its path. The available size of the field of view is therefore effectively reduced by the factor $1 - \tau v/l$ provided $\tau < l/v$, and no photon correlations at all can be contributed by a single atom to the measurement when $\tau > l/v$ or $v > l/\tau$. If $P(v)$ is the probability density of the velocity distribution of the atoms in the beam, it follows that the term $N\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle [1+\lambda_1(\tau)]$ in Eq. (16) corresponding to single-atom correlations should be multiplied by the correction factor

$$\xi(\tau) = \int_0^{l/\tau} (1 - \tau v/l) P(v)\, dv, \quad (17)$$

and the term in $|\gamma(\tau)|^2$ that involves a pair of atoms should be multiplied by $\xi^2(\tau)$. $\xi(\tau)$ is close to unity for $\tau \ll l/\langle v\rangle$, but becomes extremely small for large τ. Single-atom contributions to the correlation function are then effectively lost.

For atoms emerging in a beam through a small aperture from a reservoir in thermal equilibrium, the velocity distribution has the form

$$P(v) = 2(v^3/v_0^4)\exp(-v^2/v_0^2), \quad (18)$$

where v_0 equals $4/3\sqrt{\pi}$ times the mean velocity of the atomic beam. Hence, from Eqs. (17) and (18),

$$\xi(\tau) = 2\int_0^{l/v_0\tau} \left(1 - \frac{x v_0 \tau}{l}\right) x^3 e^{-x^2} dx, \quad (19)$$

and Eq. (16) has to be modified to become

$$\langle n_N(\tau)\rangle = \mathfrak{N}_N \Delta\tau\, \alpha_b S_b \left(N\langle \hat{I}_{1b}\rangle + \langle i_b\rangle + \frac{\xi(\tau) N \langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle [1+\lambda_1(\tau)] - N\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle + \delta_{ab}\, \xi^2(\tau)\eta N(N-1)\langle \hat{I}_{1a}\rangle \langle \hat{I}_{1b}\rangle\, |\gamma(\tau)|^2}{N\langle \hat{I}_{1a}\rangle + \langle i_a\rangle} \right). \tag{20}$$

IV. AVERAGE OVER THE ATOMIC ENSEMBLE

Eq. (20) applies to the situation in which a definite number N of atoms is in the field of view. In practice, this number fluctuates from observation to observation. There will be $\mathfrak{N}_0$ start pulses with no atoms in the field of view that produce $n_0(\tau)$ events in channel τ, $\mathfrak{N}_1$ start pulses with 1 atom producing $n_1(\tau)$ events in the same channel, etc. The apparatus registers only the total number of events

$$n(\tau) \equiv \sum_{N=0}^{\infty} n_N(\tau)\,, \tag{21}$$

and the total number of start pulses

$$\mathfrak{N} \equiv \sum_{N=0}^{\infty} \mathfrak{N}_N\,, \tag{22}$$

so that Eq. (20) has to be summed over N. Clearly $\mathfrak{N}_N$ is related to the total number $\mathfrak{N}$ by the conditional probability $p_c(N)$ that there are N atoms in the field when there is a start pulse, so that we may write

$$\mathfrak{N}_N = \mathfrak{N} p_c(N)\,. \tag{23}$$

$p_c(N)$ must be clearly distinguished from the unconditional probability $p(N)$ that there are N atoms in the field at any one time. From Eqs. (20) and (23) we then have

$$\langle n(\tau)\rangle = \mathfrak{N}\Delta\tau \sum_{N=0}^{\infty} p_c(N) \left(\frac{N}{\langle N\rangle} R_b + r_b + \frac{(NR_aR_b/\langle N\rangle^2)[\xi(\tau)+\xi(\tau)\lambda_1(\tau)-1] + \delta_{ab}\,\xi^2(\tau)\eta N(N-1)R_aR_b\,|\gamma(\tau)|^2/\langle N\rangle^2}{NR_a/\langle N\rangle + r_a} \right), \tag{24}$$

where we have introduced the mean counting rates R_a, R_b in the two channels contributed by the fluorescence

$$\begin{aligned} R_a &\equiv \alpha_a S_a \langle N\rangle \langle \hat{I}_{1a}\rangle\,, \\ R_b &\equiv \alpha_b S_b \langle N\rangle \langle \hat{I}_{1b}\rangle\,, \end{aligned} \tag{25}$$

and the mean background counting rates r_a, r_b

$$\begin{aligned} r_a &\equiv \alpha_a S_a \langle i_a\rangle\,, \\ r_b &\equiv \alpha_b S_b \langle i_b\rangle\,, \end{aligned} \tag{26}$$

all of which can be measured directly. It should be noted that the summation variable N in Eq. (24) appears both in the numerator and in the denominator, and that the denominator cannot be treated as a constant.

In order to perform the summation we need to relate the conditional probability $p_c(N)$ to the unconditional probability $p(N)$. This can be done as follows: Let $P(N, \text{start})\delta t$ be the joint probability that there are N atoms in the field and that a start pulse occurs at time t within δt. Then we obtain $p_c(N)$ by dividing the joint probability $P(N, \text{start})\delta t$ by the probability that a start pulse occurs within δt, which is $(R_a + r_a)\delta t$, or

$$p_c(N) = \frac{P(N, \text{start})\delta t}{(R_a + r_a)\delta t}\,. \tag{27}$$

To arrive at the joint probability $P(N, \text{start})\delta t$, we observe that the event in question may occur in several mutually exclusive ways, whose probabilities we can add together. There may be N atoms in the field and the start pulse may be caused by background light, which has probability $p(N)\alpha_a S_a \langle i_a\rangle \delta t$. There may be N atoms in the field, and the first one may give rise to the start pulse, which has probability $p(N)\alpha_a S_a \langle \hat{I}_{1a}\rangle \delta t$, etc. On adding all the elementary probabilities we find

$$\begin{aligned} p(N, \text{start})\delta t &= p(N)\alpha_a S_a \delta t(\langle i_a\rangle + N\langle \hat{I}_{1a}\rangle) \\ &= p(N)\delta t(r_a + NR_a/\langle N\rangle)\,, \end{aligned} \tag{28}$$

and, with the help of Eq. (27), this leads to

$$p_c(N) = p(N)\frac{r_a + (N/\langle N\rangle)R_a}{r_a + R_a}\,. \tag{29}$$

When this result is inserted in Eq. (24), we finally obtain

$$\langle n(\tau)\rangle = \mathfrak{N}\Delta\tau\left\{r_b + R_b + \frac{R_a R_b}{R_a + r_a}\left[\frac{\xi(\tau)}{\langle N\rangle}[1+\lambda_1(\tau)] + \frac{\langle(\Delta N)^2\rangle - \langle N\rangle}{\langle N\rangle^2} + \delta_{ab}\xi^2(\tau)\eta|\gamma(\tau)|^2\left(1 + \frac{\langle(\Delta N)^2\rangle - \langle N\rangle}{\langle N\rangle^2}\right)\right]\right\}, \quad (30)$$

which reduces further, with the assumption of a Poissonian form for the atomic distribution $p(N)$, to

$$\langle n(\tau)\rangle = \mathfrak{N}\Delta\tau\left\{r_b + R_b + \left(\frac{R_a R_b}{R_a + r_a}\right)\left[\frac{\xi(\tau)}{\langle N\rangle}[1+\lambda_1(\tau)] + \delta_{ab}\xi^2(\tau)\eta|\gamma(\tau)|^2\right]\right\}. \quad (31)$$

This formula differs from the one quoted in Ref. 1 in several respects, for reasons that we have mentioned. One interesting conclusion is that, when two photodetectors are used so that the $|\gamma(\tau)|^2$ term drops out and when $\tau = 0$, so that $\lambda_1(0) = -1$, the expected number of events $\langle n(0)\rangle$ in channel 0 is just $\mathfrak{N}\Delta\tau(R_b + r_b)$. This is also the number that is expected in the absence of photon correlations of any kind. As τ increases, $\langle n(\tau)\rangle$ also increases, reaches a maximum, and returns to the initial value $\mathfrak{N}\Delta\tau(R_b + r_b)$ for sufficiently large τ. This behavior was in fact observed in the experiment.[1] Except for the factor $\xi(\tau)$, the $[1+\lambda_1(\tau)]/\langle N\rangle$ term in Eq. (31) was also obtained by Jakeman *et al.*,[5] although they used a somewhat oversimplified argument to derive it.[10]

As the average number of atoms $\langle N\rangle$ in the field of view becomes smaller, the $[1+\lambda_1(\tau)]/\langle N\rangle$ term in Eq. (31) becomes the dominant term (except near $\tau = 0$), provided the background is small enough, and the equation reduces to the usual form for a single atom.[2,3] On the other hand, when $\langle N\rangle$ is very large, the $[1+\lambda_1(\tau)]/\langle N\rangle$ term becomes small, and the antibunching becomes less and less evident. This is particularly true when a single photodetector is used and the $|\gamma(\tau)|^2$ term, which always has its greatest value when $\tau = 0$, makes a significant contribution to Eq. (31). The importance of using very feeble atomic beams in photon antibunching measurements is therefore clearly brought out.

V. COMPARISON WITH EXPERIMENT

Figure 1 shows a comparison between the prediction of Eq. (31) and some experimental results obtained in a photoelectric-correlation measurement of the type reported in Ref. 1 of the fluorescence from the $3P_{3/2}$, $F = 3$, $m_F = 3$ to $3^2S_{1/2}$, $F = 2$, $m_F = 2$ transition in sodium. Two photodetectors were used in the experiment, so that $\delta_{ab} = 0$ in Eq. (31). The exciting light was on resonance and had an intensity equivalent to 10 ± 2 mW/cm^2, corresponding to a relative Rabi frequency $\Omega/\beta \approx 1.7 \pm 0.2$ in the notation of Ref. 3. The expected form of $\lambda_1(\tau)$ under these conditions was taken from Ref. 3. Two sets of measurements were carried out at two different atomic-beam currents, with the same exciting light. In the first, corresponding to the upper set of points in Fig. 1, the mean number of atoms $\langle N\rangle$ in the field of view was estimated from the measured fluorescence to be about $\frac{1}{3}$ with an uncertainty of 50%. In the second set of measurements, corresponding to the lower set of points, the atomic-beam current was increased by a factor 2.7 ± 0.1 as determined from the fluorescence, although the light intensity at each detector was correspondingly reduced by a filter, to keep the counting rates approximately the same. The rates were $R_a = 15\,818$ sec^{-1}, $R_b = 16\,796$ sec^{-1}, $r_a = 538$ sec^{-1}, $r_b = 472$ sec^{-1} in the first experiment and $R_a = 13\,323$ sec^{-1}, $R_b = 13\,752$ sec^{-1}, $r_a = 246$ sec^{-1}, and $r_b = 223$ sec^{-1} in the second. The number $\mathfrak{N}$ of start pulses was 9×10^6 in both cases, and the time interval τ is shown digitized in steps of $\Delta\tau = 2.5$ nsec, although it was actually measured in $\frac{1}{2}$-nsec intervals. The average transit time of the atoms through the field of view was estimated to be 100 ± 15 nsec, as

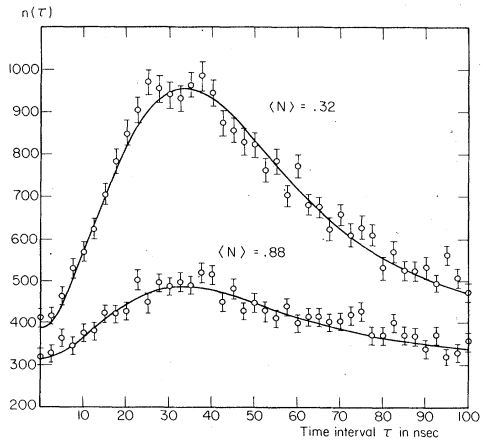

FIG. 1. Comparison of the values of $n(\tau)$ measured in a photon-correlation experiment with the theoretical values for $\langle n(\tau)\rangle$ given by Eq. (31) with $\Omega/\beta = 1.75$, for two sets of measurements with different atomic-beam currents. The initial rise of $n(\tau)$ from $\tau = 0$ is evidence for photon antibunching.

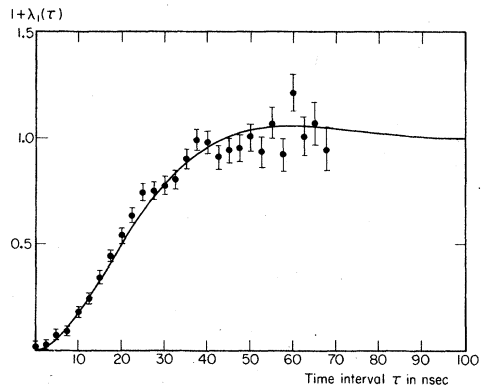

FIG. 2. Comparison of the values of the correlation function $1+\lambda_1(\tau)$ derived from the measurements with those given by the theory (full curve) with $\Omega/\beta=1.75$. The statistical uncertainties get progressively larger as τ increases.

determined from the temperature of the exit aperture of the atomic-beam oven. The full curves in Fig. 1 are calculated from Eq. (31) with $\langle N\rangle=0.32$, $l/v_0=105$ nsec and $\langle N\rangle=0.88$, $l/v_0=105$ nsec. It will be seen that there is very good agreement between theory and experiment within the statistical uncertainties, so that the validity of Eq. (31) is confirmed at least for measurements with two detectors. Moreover, the importance of keeping $\langle N\rangle$ small if antibunching is to be observed is clearly brought out. It has recently been pointed out by Agarwal *et al.*[11] that cooperative effects between two or more atoms would also greatly reduce the antibunching, although we do not believe that such effects were significant in these experiments.

However, the strong attenuation imposed by the transit time factor $\xi(\tau)$ in Eq. (31) for the larger delays τ makes it difficult to extract the correlation function $\lambda_1(\tau)$ accurately from the measured values of $n(\tau)$. This is illustrated in Fig. 2, where the values of $1+\lambda_1(\tau)$ derived from the upper set of data points in Fig. 1, together with their standard deviations, are shown superimposed on the theoretical curve. As τ increases and the measured values of $n(\tau)$ approach $\mathfrak{N}\Delta\tau(R_b+r_b)$, the relative statistical uncertainties become progressively larger, until the data will fit almost any curve. With the atomic-transit time l/v_0 of order 100 nsec, the derived values are hardly meaningful for τ greater than about 50 nsec. This brings out the importance of not making the transit time, and therefore the field of view, too small. In practice, as always, a compromise has to be reached between choosing a sufficiently large field of view and keeping the background light intensity sufficiently small.

ACKNOWLEDGMENT

The work at the University of Rochester was supported in part by the National Science Foundation. One of the authors (M.D.) wishes to thank the National Research Council for financial support.

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