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Photoelectron waiting times and atomic state reduction in resonance fluorescence

H. J. Carmichael, Surendra Singh, Reeta Vyas, and P. R. Rice*
Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701
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Photoelectron counting sequences for single-atom resonance fluorescence are studied. The distribution of waiting times between photoelectric counts is calculated, and its dependence on driving-field intensity and detection efficiency is discussed. The photoelectron-counting distribution is derived from the waiting-time distribution. The relationship between photoelectron counting sequences and photon emission sequences is discussed and used to obtain an expression for the reduced state of the atom during the waiting times between photoelectric counts. The roles of irreversibility and the observer in atomic state reduction are delineated.

I. INTRODUCTION

The fluorescent photons emitted by a single coherently driven two-level atom exhibit the nonclassical property of photon antibunching.[1-3] The antibunching of fluorescent photons is seen in temporal correlations between photoelectric counts; the detection of one photon makes the detection of a second, after just a short delay, improbable. Photon antibunching is traditionally defined in terms of the degree of second-order temporal coherence $g^{(2)}(t,t+\tau)$. This is the joint probability for recording photoelectric counts in the intervals $[t,t+\Delta t)$ and $[t+\tau,t+\tau+\Delta t)$, normalized by the probability for two independent photoelectric counts. For antibunched light the joint probability for recording photoelectric counts closely spaced in time falls below the probability for statistically independent counts (separated by a time longer than the coherence time); thus, $g^{(2)}(t,t)<1$.

The antibunching of fluorescent photons is also reflected in the sub-Poissonian character of the probability density $p(n,t,t+T)$ for recording n photoelectric counts in the interval $[t,t+T)$.[4] $p(n,t,t+T)$ can be derived from $g^{(2)}(t,t+\tau)$, although the detailed algebraic relationship is quite complicated. Both $g^{(2)}(t,t+\tau)$ and $p(n,t,t+T)$ have been calculated for single-atom resonance fluorescence by a number of workers.[1-10] Because of the complexity of general expressions in the time domain, some workers only give the Laplace transform for the photoelectron counting distribution, or give explicit time-dependent expressions only for limiting cases, such as short and long counting times.

Recent theoretical work on "quantum jumps"[11,12] has drawn attention to the distribution of waiting times between photon emissions as another useful quantity for characterizing photon statistics—in terms of measured quantities, the distribution of waiting times between photoelectrons. By "waiting time" we mean the time τ between a photoelectric count recorded at time t, and the next, recorded at time $t+\tau$. If photoelectron sequences can be described by a Markov birth process, a single conditional probability density $w(\tau|t)$ specifies the distribution of waiting times between every pair of photoelectrons. We call this the photoelectron waiting-time distribution. Photoelectron waiting times for coherent light are exponentially distributed.[13] Antibunching implies that photons tend to be separated in time. The distribution of waiting times should then tend to peak around the average time between photoelectric counts.

Photoelectron waiting times are certainly not new to the field of photon statistics. Indeed, when a time-to-amplitude converter is used for a delayed coincidence measurement, the raw data provide the distribution of waiting times between photoelectric counts. However, when the count rate is sufficiently low, this distribution is proportional (aside from dead-time corrections) to $g^{(2)}(t,t+\tau)$.[14] This relationship provides the technique used to measure $g^{(2)}(t,t+\tau)$ in the experiments of Kimble *et al.*[3] on photon antibunching in resonance fluorescence. Thus, the waiting-time distribution and its relationship to $g^{(2)}(t,t+\tau)$ are known. But the waiting-time distribution has not been mentioned until recently[12,15,16] in the large theoretical literature on resonance fluorescence. This is a deficiency, since it provides a clearer physical picture of photon emission sequences, corresponding photoelectron counting sequences, and their nonclassical properties, than $g^{(2)}(t,t+\tau)$. In this paper we revisit the problem of single-atom resonance fluorescence and focus attention on the waiting-time distribution. (We will discuss waiting times between photon emissions as well as between photoelectrons. When the distinction is not important we simply refer to "the waiting times" or "the waiting-time distribution.")

There are probably two main reasons for the lack of attention paid to $w(\tau|t)$ in early work on resonance fluorescence. The first is that $g^{(2)}(t,t+\tau)$, not $w(\tau|t)$, is the quantity accessible to measurement. It might be asked, why not use a time-to-amplitude converter to measure the quantity it gives directly, the photoelectron waiting-time distribution $w(\tau|t)$? The problem is that photoelectric detection is very inefficient. The average time between photoelectric counts is unavoidably much longer than the correlation time of the fluorescent light. Then $w(\tau|t)$ is proportional to $g^{(2)}(t,t+\tau)$; $w(\tau|t)$ can be measured, but only when it effectively reduces to $g^{(2)}(t,t+\tau)$. Actually, the proportionality between these quantities does not hold for all τ, but it holds over many correlation

times, beyond which $w(\tau|t)$ decays at a rate proportional to the detection efficiency.[14–16]

The second reason for the lack of attention paid to $w(\tau|t)$ concerns the theory of photoelectric counting, which focuses attention on *nonexclusive* probabilities, and tends to neglect the *exclusive* probabilities which are related to $w(\tau|t)$. The Kelly-Kleiner treatment of photoelectric counting is formulated in terms of the nonexclusive probability densities, or multicoincidence rates, $w_m(t_1,t_2,\ldots,t_m)$;[17] $w_m(t_1,t_2,\ldots,t_m)\Delta t_1\Delta t_2\cdots\Delta t_m$ is the probability that one photoelectric count is recorded in each of the nonoverlapping intervals $(t_1<t_2<\cdots<t_m)$

$$[t_1,t_1+\Delta t_1),[t_2,t_2+\Delta t_2),\ldots,[t_m,t_m+\Delta t_m) .$$

No restriction is placed on the number of counts recorded outside these intervals. The hierarchy of field correlation functions introduced by Glauber[18] is defined in terms of these nonexclusive probabilities. In particular, the degree of second-order coherence is given by

$$g^{(2)}(t,t+\tau)=w_2(t,t+\tau)/[w_1(t)w_1(t+\tau)] . \tag{1}$$

Quantities of experimental interest are naturally expressed in terms of nonexclusive probabilities: of course, the multicoincidence counting rates, and also factorial moments of the photoelectron counting distribution.[17]

Stochastic photoelectric counting sequences can also be characterized by the hierarchy of exclusive probability densities $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$;[13,17]

$$\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])\Delta t_1\Delta t_2\cdots\Delta t_m$$

is the probability that m photoelectric counts are recorded in the observation interval $[t,t+T]$, one in each of the nonoverlapping intervals $(t_1<t_2<\cdots<t_m)$

$$[t_1,t_1+\Delta t_1),[t_2,t_2+\Delta t_2),\ldots,[t_m,t_m+\Delta t_m) .$$

The waiting-time distribution is related to the conditional probability densities

$$\wp_m(t_1,t_2,\ldots,t_m|t_0)$$

$$=\wp_{m+1}(t_0,t_1,t_2,\ldots,t_m;[t_0,t_m])/w_1(t_0) ; \tag{2}$$

$\wp_m(t_1,t_2,\ldots,t_m|t_0)\Delta t_1\Delta t_2\cdots\Delta t_m$ is the probability that, given a photoelectric count is recorded at t_0, the next m photoelectric counts are recorded in the nonoverlapping intervals $(t_1<t_2<\cdots<t_m)$

$$[t_1,t_1+\Delta t_1),[t_2,t_2+\Delta t_2),\ldots,[t_m,t_m+\Delta t_m) .$$

For Markov counting sequences

$$\wp_m(t_1,t_2,\ldots,t_m|t_0)=\prod_{i=1}^{m}\wp_1(t_i|t_{i-1}) . \tag{3}$$

The distribution of waiting times τ between a photoelectric count recorded at time t, and the next, recorded at $t+\tau$, is given by

$$w(\tau|t)\equiv\wp_1(t+\tau|t) . \tag{4}$$

The exclusive probabilities are not naturally related to measureable quantities. However, they provide the elementary probabilities in terms of which $p(n,t,t+T)$ is most naturally defined:

$$p(n,t,t+T)=\int_t^{t+T}dt_n\int_t^{t_n}dt_{n-1}\cdots\int_t^{t_2}dt_1\wp_n(t_1,t_2,\ldots,t_n;[t,t+T]) ; \tag{5}$$

for Markov counting sequences

$$\wp_n(t_1,t_2,\ldots,t_n;[t,t+T])=\frac{\wp_1(t_n;[t_n,t+T])}{w_1(t_n)}\left[\prod_{i=2}^{n}w(t_i-t_{i-1}|t_{i-1})\right]\wp_1(t_1;[t,t_1]) , \tag{6}$$

where $\wp_1(t_1;[t,t_1])\Delta t_1$ is the probability that, if observation begins at t, the first photoelectric count is recorded in the interval $[t_1,t_1+\Delta t_1)$, and $\wp_1(t_n;[t_n,t+T])/w_1(t_n)$ is the probability that, given a photoelectric count is recorded at t_n, no photoelectric counts are recorded in the interval $(t_n,t+T]$. The photoelectron waiting-time distribution determines the intervals between pairs of successive photoelectric counts, and is therefore a quantity around which we can build a strong conceptual understanding of the counting sequences. Of course, if photoelectron counting sequences are not Markovian $w(\tau|t)$ is not so central; it is then just one of the hierarchy of probability densities $\wp_m(t_1,t_2,\ldots,t_m|t_0)$.

The existing derivations of $p(n,t,t+T)$ for resonance fluorescence fall into two categories. Most use the Kelly-Kleiner formula expressed in terms of nonexclusive probabilities.[4,7–10] However, two derivations directly track sequences of photon emissions from the atomic source.[5,6] These effectively calculate $p(n,t,t+T)$ from exclusive probabilities, as in Eq. (6), although they do not use this language explicitly. In this paper we provide the formal connection between treatments of photon statistics based on the standard Kelly-Kleiner formulation, and treatments made in terms of dynamical equations for the emitting source. Lenstra[8] showed that Cook's derivation of $p(n,t,t+T)$ (Ref. 6) gives the same results as the Kelly-Kleiner formula. Cook's treatment seems rather removed from the theory of photoelectron counting since it is based on the equations for momentum transfer from the driving field to the atom. It is, however, effectively a

derivation (for perfect detection efficiency) from Eq. (5). We show the relationship between source emissions and the Kelly-Kleiner theory in general formal terms, allowing for arbitrary detection efficiency.

This formal relationship gives a clear picture of the role of the detector in atomic state reduction. The detector is not the agent *causing* state reduction. The atom collapses to its ground state due to its *irreversible* decay into the vacuum; the collapses proceed at an average rate given by the inverse atomic lifetime, quite indifferent to the successful or unsuccessful recording of the emitted photons. Photoelectric detection monitors these photons with greater or lesser precision depending on the detection efficiency. We calculate the time development of the reduced atomic state *inferred* by an observer during the waiting time between photoelectric counts. This state depends on detection efficiency. It reflects the observer's lack of knowledge about the continuing collapses of the atomic state associated with undetected photon emissions.

Our plan is to attack the problem of photoelectron counting statistics in resonance fluorescence from two directions, and to relate the two approaches. Sections II–IV provide a development from the standard Kelly-Kleiner theory, then in Sec. V we show how the same results can be obtained from the dissipative dynamics of the source. In Sec. II we derive the photoelectron waiting-time distribution from its relationship to the nonexclusive probability densities $w_m(t_1,t_2,\ldots,t_m)$. We discuss the dependence of the waiting-time distribution on the driving-field strength and detection efficiency, and show how the nonclassical properties of photon antibunching and sub-Poissonian statistics are clearly reflected in $w(\tau|t)$. In Sec. III we show how standard expressions for $w(\tau|t)$, and more generally $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$, can be reexpressed so that these quantities can be derived directly from modified Bloch equations for the atom. We derive a formal expression for exclusive probability densities from which we identify the reduced state of the atom inferred during the waiting times between photoelectric counts. In Sec. IV we calculate the photoelectron counting distribution from Eq. (5). In Sec. V we derive the same expression for exclusive probability densities without reference to the theory of photoelectric detection, starting from the dynamical equations for the atomic source. We describe the formal connection between photon emission sequences and photoelectron counting sequences and discuss the issue of atomic state reduction. Section VI provides a summary and conclusions.

II. THE PHOTOELECTRON WAITING-TIME DISTRIBUTION

The fluorescent light from a coherently driven two-level atom is incident on the photocathode of a photoelectric detector. The detector quantum efficiency is η $(0<\eta\leq 1)$, and the quasimonochromatic field at the detector is described by the photon-flux operator $\hat{I}(t)\propto\hat{E}^{(-)}(t)\hat{E}^{(+)}(t)$, where $\hat{E}^{(+)}(t)$ and $\hat{E}^{(-)}(t)$ are the positive and negative frequency components of the electric field [the mean photoelectron counting rate is $\eta\langle\hat{I}(t)\rangle$]. The nonexclusive probability densities $w_m(t_1,t_2,\ldots,t_m)$ are given in terms of η and $\hat{I}(t)$ by[17,18]

$$w_m(t_1,t_2,\ldots,t_m)=\eta^m\langle\mathcal{T}{:}\hat{I}(t_m)\cdots\hat{I}(t_2)\hat{I}(t_1){:}\rangle\ ; \tag{7}$$

:: stands for normal ordering of field operators, and $\mathcal{T}$ means that operators $\hat{E}^{(-)}$ and $\hat{E}^{(+)}$ are to be written in ascending and descending time order, respectively. The exclusive probability densities $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ can be calculated from the nonexclusive probabilities:[13]

$$\begin{aligned}\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])&=\eta^m\sum_{r=0}^{\infty}\frac{(-\eta)^r}{r!}\int_t^{t+T}dt'_r\int_t^{t+T}dt'_{r-1}\cdots\int_t^{t+T}dt'_1\langle\mathcal{T}{:}\hat{I}(t'_r)\hat{I}(t'_{r-1})\cdots\\&\qquad\times\hat{I}(t'_1)\hat{I}(t_m)\cdots\hat{I}(t_2)\hat{I}(t_1){:}\rangle\\&=\eta^m\langle\mathcal{T}{:}\exp[-\hat{\Omega}(t+T,t)]\hat{I}(t_m)\cdots\hat{I}(t_2)\hat{I}(t_1){:}\rangle\\&=\eta^m\langle\mathcal{T}{:}\exp[-\hat{\Omega}(t+T,t_m)]\hat{I}(t_m)\cdots\hat{I}(t_2)\exp[-\hat{\Omega}(t_2,t_1)]\hat{I}(t_1)\exp[-\hat{\Omega}(t_1,t)]{:}\rangle\ ,\end{aligned} \tag{8}$$

where

$$\hat{\Omega}(t_i,t_j)=\eta\int_{t_j}^{t_i}dt'\hat{I}(t')\ . \tag{9}$$

Then, from Eq. (2), the conditional probability densities $\wp_m(t_1,t_2,\ldots,t_m|t_0)$ are given by

$$\wp_m(t_1,t_2,\ldots,t_m|t_0)=\langle\hat{I}(t)\rangle^{-1}\eta^m\langle\mathcal{T}{:}\hat{I}(t_m)\exp[-\hat{\Omega}(t_m,t_{m-1})]\cdots\hat{I}(t_2)\exp[-\hat{\Omega}(t_2,t_1)]\hat{I}(t_1)\exp[-\hat{\Omega}(t_1,t_0)]\hat{I}(t_0){:}\rangle. \tag{10}$$

Single-atom resonance fluorescence yields Markov photoelectron counting sequences; therefore, $\wp_m(t_1,t_2,\ldots,t_m|t_0)$ and $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ factorize as in Eqs. (3) and (6). The formal proof of this factorization is left until the next section. However, the Markov property is easily appreciated on physical grounds. It follows from the argument that is used to explain the antibunching of fluorescent photons. This states that when each photoelectric count is recorded, the atom is known to have returned to its ground state with

the emission of one photon. Thus, after each photoelectric count, the atom always evolves from the same state—its ground state; therefore, the subsequent state of the atom does not depend in any way on the history of counts previously recorded. The probability for recording the ith photoelectric count in the time interval $[t_i, t_i+\Delta t_i)$ will then depend on the time t_{i-1} of the last photoelectric count, but it will not depend on the history of prior photoelectric counts. Photoelectron counting sequences are therefore Markovian.

The return of the atom to its ground state when a photoelectric count is recorded also guarantees that photoelectron counting sequences are stationary. More precisely, they are stationary for all but the very first count. The conditional probability densities $\wp_m(t_1,t_2,\ldots,t_m|t_0)$ are independent of the time t_0. This is true even if the atom is not in a stationary state prior to the count at t_0. We therefore define

$$\wp_m(\tau_1,\tau_2,\ldots,\tau_m)\equiv\wp_m(t_0+\tau_1,t_0+\tau_2,\ldots,t_0+\tau_m|t_0)\ , \tag{11}$$

and write Eq. (10) in the form

$$\wp_m(\tau_1,\tau_2,\ldots,\tau_m)$$
$$=\langle\hat{I}_0(0)\rangle^{-1}\eta^m\langle\mathcal{T}{:}\hat{I}_0(\tau_m)\exp[-\hat{\Omega}_0(\tau_m,\tau_{m-1})]\cdots\hat{I}_0(\tau_2)\exp[-\hat{\Omega}_0(\tau_2,\tau_1)]\hat{I}_0(\tau_1)\exp[-\hat{\Omega}_0(\tau_1,0)]\hat{I}_0(0){:}\rangle\ , \tag{12}$$

with

$$\hat{\Omega}_0(\tau_i,\tau_j)=\eta\int_{\tau_j}^{\tau_i}dt'\hat{I}_0(t')\ , \tag{13}$$

where

$$\hat{I}_0(\tau)\equiv\hat{I}(t_0+\tau)\ , \tag{14}$$

and t_0 is an arbitrary time (allowing for retardation) after excitation of the atom begins. We define the waiting-time distribution by

$$w(\tau)\equiv w(\tau|t_0)\equiv\wp_1(t_0+\tau|t_0)\ . \tag{15}$$

From the Markov property, and Eqs. (12) and (15),

$$\wp_m(\tau_1,\tau_2,\ldots,\tau_m)=\prod_{i=1}^{m}w(\tau_i)\ , \tag{16}$$

where

$$w(\tau)=\langle\hat{I}_0(0)\rangle^{-1}\eta\langle\mathcal{T}{:}\hat{I}_0(\tau)\exp[-\hat{\Omega}_0(\tau,0)]\hat{I}_0(0){:}\rangle\ . \tag{17}$$

In this section we calculate $w(\tau)$ from Eq. (17).

We first note the connection between the photoelectron waiting-time distribution and the degree of second-order temporal coherence. For times that are much less than the average time $(\eta\langle\hat{I}\rangle)^{-1}$ between photoelectric counts, exclusive probabilities are simply related to nonexclusive probabilities. We may replace the exponentials in Eqs. (8), (12), and (17) by unity, and write $[t_i-t_{i-1}\ll(\eta\langle\hat{I}\rangle)^{-1}]$

$$\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])=w_m(t_1,t_2,\ldots,t_m)\ , \tag{18}$$

$$\wp_m(\tau_1,\tau_2,\ldots,\tau_m)=w_{m+1}(t_0,t_0+\tau_1,t_0+\tau_2,\ldots,t_0+\tau_m)/w_1(t_0)\ , \tag{19}$$

and $[\tau\ll(\eta\langle\hat{I}\rangle)^{-1}]$

$$w(\tau)=w_2(t_0,t_0+\tau)/w_1(t_0)=\eta\langle\hat{I}\rangle_{ss}g_{ss}^{(2)}(\tau)\ ; \tag{20}$$

$\eta\langle\hat{I}\rangle_{ss}$ and $g_{ss}^{(2)}(\tau)$ are the mean photoelectron counting rate and the degree of second-order temporal coherence in the steady-state limit $t_0\to\infty$. When either the detection efficiency or the photon flux is sufficiently small, the inequality $\tau\ll(\eta\langle\hat{I}\rangle)^{-1}$ will hold over many correlation times. Under these conditions $g_{ss}^{(2)}(\tau)$ can be obtained directly from a measurement of $w(\tau)$ using a time-to-amplitude converter.[2,14] The relationships expressed by Eqs. (18)–(20) are easily understood. For short time intervals t_i-t_{i-1} between photoelectric counts, the probability for recording other counts during these intervals is vanishingly small. Therefore, the probability density for recording photoelectric counts at $t_1=t_0+\tau_1$, $t_2=t_0+\tau_2,\ldots,t_m=t_0+\tau_m$ is the same, whether or not it is conditioned on the requirement that no counts be recorded at other times.

To calculate $w(\tau)$ for times that are not short compared to the average time between photoelectric counts, we expand the exponent in Eq. (17) and write

$$w(\tau)=\langle\hat{I}_0(0)\rangle^{-1}\eta\sum_{m=0}^{\infty}(-\eta)^m\int_0^{\tau}d\tau_m\int_0^{\tau_m}d\tau_{m-1}\cdots\int_0^{\tau_2}d\tau_1\langle\mathcal{T}{:}\hat{I}_0(\tau)\hat{I}_0(\tau_m)\hat{I}_0(\tau_{m-1})\cdots\hat{I}_0(\tau_1)\hat{I}_0(0){:}\rangle\ . \tag{21}$$

The correlation functions that appear inside the integrals are proportional to the nonexclusive probability densities

$$w_{m+2}(t_0,t_0+\tau_1,t_0+\tau_2,\ldots,t_0+\tau_m,t_0+\tau)\ .$$

These correlation functions factorize by the Markov property (a formal proof is given in Ref. 19); we have

$$\langle \mathcal{T}:\hat{I}_0(\tau)\hat{I}_0(\tau_m)\hat{I}_0(\tau_{m-1})\cdots\hat{I}_0(\tau_1)\hat{I}_0(0):\rangle = f_0(\tau-\tau_m)f_0(\tau_m-\tau_{m-1})\cdots f_0(\tau_1)\langle\hat{I}_0(0)\rangle \ , \tag{22}$$

where [read $f(\tau_1)=f(\tau_1-0)$]

$$\begin{aligned}\eta f_0(\tau_i-\tau_{i-1}) &= \eta\langle\hat{I}_0(\tau_i)\rangle g^{(2)}(t_0+\tau_i,t_0+\tau_{i-1}) \\ &= \eta\langle\hat{I}\rangle_{ss} g^{(2)}_{ss}(\tau_i-\tau_{i-1})\end{aligned} \tag{23}$$

is the conditional probability density for recording a photoelectric count at time $t_0+\tau_i$, given that a count was recorded at time $t_0+\tau_{i-1}$; this probability density is independent of t_0 and we therefore express it in terms of steady-state quantities ($t_0\to\infty$). By substituting Eq. (22) into Eq. (21), we have

$$w(\tau)=\eta\sum_{m=0}^{\infty}(-\eta)^m\int_0^{\tau}d\tau_m\int_0^{\tau_m}d\tau_{m-1}\cdots\int_0^{\tau_2}d\tau_1 f_0(\tau-\tau_m)f_0(\tau_m-\tau_{m-1})\cdots f_0(\tau_1) \ . \tag{24}$$

The infinite series can now be summed by taking Laplace transforms with respect to τ. We find[20]

$$\tilde{w}(s)=\frac{\eta\tilde{f}_0(s)}{1+\eta\tilde{f}_0(s)} \ , \tag{25}$$

where $\tilde{w}(s)$ and $\tilde{f}_0(s)$ are the Laplace transforms of $w(\tau)$ and $f_0(\tau)$, respectively.

The detailed dynamics of the atomic scattering process enter through the explicit form of $f_0(\tau)$. Using Eq. (23), and familiar results for $\langle\hat{I}\rangle_{ss}$ and $g^{(2)}_{ss}(\tau)$,[1,2] we have

$$\tilde{f}_0(s)=\xi(2\beta)\frac{1}{2s}\frac{\Omega^2}{(s+2\beta)(s+\beta)+\Omega^2} \ . \tag{26}$$

2β is the Einstein A coefficient, Ω is the Rabi frequency, and ξ ($0<\xi\le 1$) is the collection efficiency of the detection system. We define an overall detection efficiency $\eta'=\eta\xi$, and then

$$\tilde{w}(s)=\frac{\eta'\beta\Omega^2}{s(s+2\beta)(s+\beta)+\Omega^2(s+\eta'\beta)} \ . \tag{27}$$

The inverse Laplace transform gives

$$w(\tau)=\sum_{i=1}^{3}[(s-s_i)\tilde{w}(s)]_{s=s_i}\exp(s_i\tau) \ , \tag{28}$$

where s_1, s_2, and s_3 are the roots of the cubic equation

$$s(s+2\beta)(s+\beta)+\Omega^2(s+\eta'\beta)=0 \ . \tag{29}$$

The photoelectron waiting-time distribution provides a clear picture of the antibunched and sub-Poissonian character of the photoelectron counting sequences. We discuss these features first for counting sequences produced with unit detection efficiency. When $\eta'=1$, every emitted photon is recorded as a photoelectric count. Then the statistics of the photon emissions are reflected, uncontaminated by random deletions, in the statistics of the photoelectric counts.

Unit detection efficiency (waiting times for photon emissions)

By setting $\eta'=1$ in Eq. (29), we find $s_1=-\beta$, and $s_{2,3}=-\beta\pm(\beta^2-\Omega^2)^{1/2}$. The waiting-time distribution is given by

$$w(\tau)=\frac{2\beta\Omega^2}{\beta^2-\Omega^2}e^{-\beta\tau}\sinh^2[\tfrac{1}{2}(\beta^2-\Omega^2)^{1/2}\tau] \ . \tag{30}$$

The average waiting time is

$$\bar{\tau}=\int_0^{\infty}d\tau\,\tau w(\tau)=\langle\hat{I}\rangle_{ss}^{-1} \ , \tag{31}$$

where

$$\langle\hat{I}\rangle_{ss}=(2\beta)\frac{1}{2}\frac{\Omega^2}{\Omega^2+2\beta^2} \tag{32}$$

is the average steady-state photoelectron counting rate; $\langle\hat{I}\rangle_{ss}$ is the product of the Einstein A coefficient and the probability for the atom to be found in its excited state.

The distribution of τ about its mean $\bar{\tau}$ reveals the nonclassical character of the fluorescent light. In Fig. 1 we compare the distribution given by Eq. (30) with that for photoelectron counting sequences with the same $\bar{\tau}$ produced by coherent light. Coherent light produces uncorrelated photoelectric counts. The waiting-time distribution can be obtained from Eq. (25), setting $\eta f_0(s)=s/\bar{\tau}$, corresponding to $g^{(2)}_{ss}(\tau)=1$; waiting times are exponentially distributed:

$$w_{\rm coh}(\tau)=(1/\bar{\tau})e^{-\tau/\bar{\tau}} \ . \tag{33}$$

For coherent light $w_{\rm coh}(0)$ is a maximum. Thus, the most probable waiting times are short waiting times. In comparison, each of the curves in Fig. 1 has $w(0)=0$. Short waiting times are therefore improbable in photoelectron counting sequences for resonance fluorescence. This comparison identifies the antibunching of photoelectric counts. Indeed, since $w(\tau)$ is proportional to $g^{(2)}_{ss}(\tau)$ for $\tau\ll\bar{\tau}$ [Eq. (20)], the condition $w(0)<w_{\rm coh}(0)=(1/\bar{\tau})$ is equivalent to the condition $g^{(2)}_{ss}(0)<1$.

The full waiting-time distribution gives a more complete picture. The integral of $w(\tau)$ over all τ is unity. Therefore, if $w(\tau)$ falls below $w_{\rm coh}(\tau)$ for short times, it must rise above $w_{\rm coh}(\tau)$ for longer times. However, we are comparing distributions that have the same mean. It is not possible then for the curves $w(\tau)$ and $w_{\rm coh}(\tau)$ to cross only once. They must cross at least twice. Figure 1(b) gives a good example. The waiting-time distribution

in this figure describes photoelectron counting sequences that are quite explicitly *antibunched* in comparison with a random counting sequence (with the same $1/\bar{\tau}$). Both short *and long* waiting times are less likely than for random counts; waiting times close to the average, $\bar{\tau}$, are more likely. The result is a peaked distribution that reveals a tendency for photoelectric counts to be recorded at regularly spaced times.

The condition $g_{ss}^{(2)}(0)<1$ identifies the existence of such a global redistribution of waiting times. But, on its own, $g_{ss}^{(2)}(0)-1$ is a poor measure of how much photoelectron counting sequences deviate from those for coherent light. Figure 1(a) shows the form of $w(\tau)$ for weak driving fields ($\Omega^2/\beta^2 \ll 1$). In this limit we find

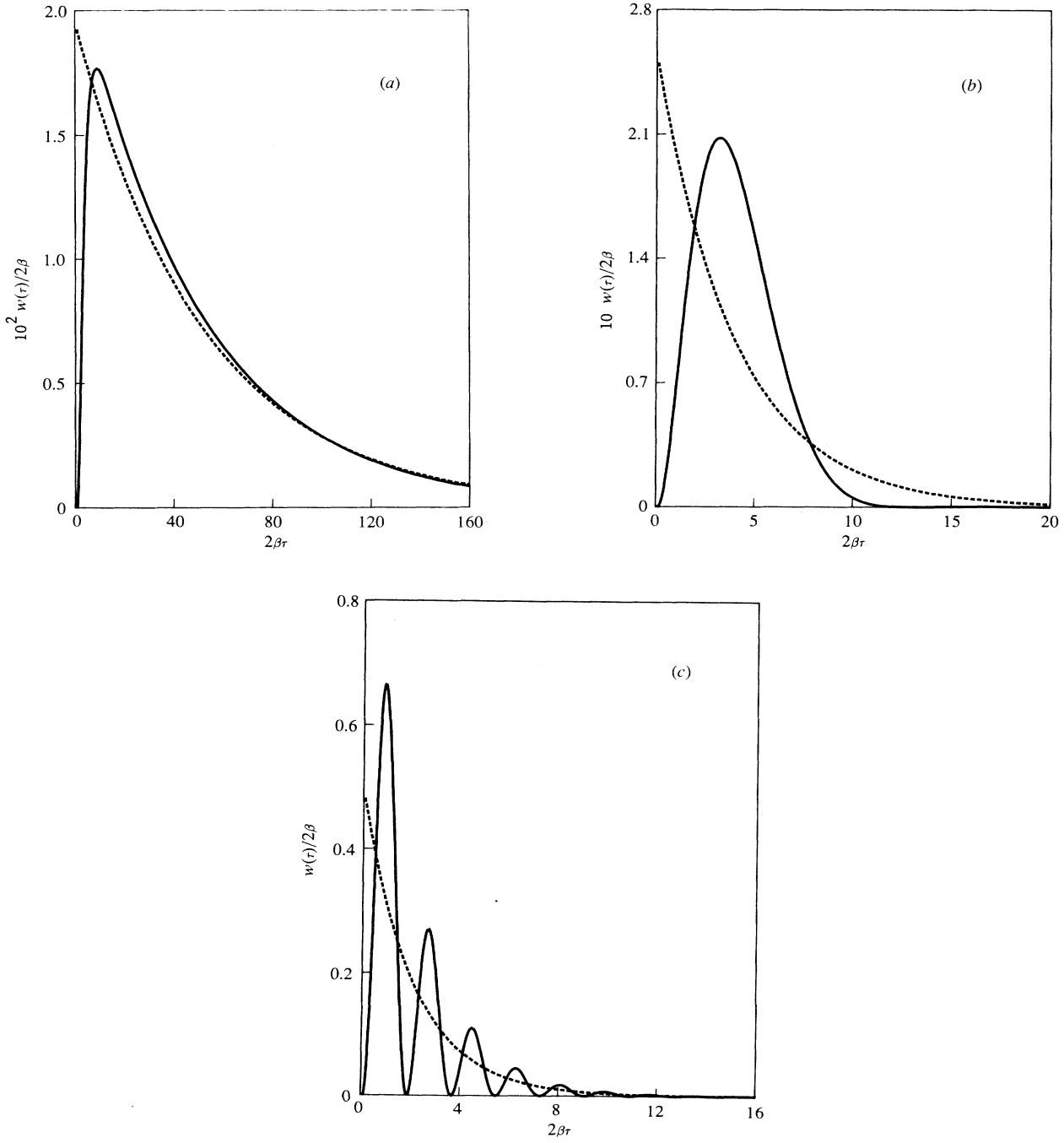

FIG. 1. Comparison of the photoelectron waiting-time distributions for resonance fluorescence and coherent light of equal intensity: $\eta'=1$, and (a) $\Omega/\sqrt{2}\beta=0.2$, (b) $\Omega/\sqrt{2}\beta=1$, (c) $\Omega/\sqrt{2}\beta=5$.

$$w(\tau)=\beta y(1+2y)\{\exp[-y(1+y/2)\beta\tau]-e^{-\beta\tau}(2-e^{\beta\tau})\}, \tag{34}$$

and

$$w_{\rm coh}(\tau)=\beta y(1-y)e^{-y(1-y)\beta\tau}, \tag{35}$$

where

$$y=\Omega^2/2\beta^2 . \tag{36}$$

We have kept terms in y to sufficient order that $\int_0^\infty d\tau\, w(\tau)=1+O(y^2)$. This is necessary to give an accurate comparison between $w(\tau)$ and $w_{\rm coh}(\tau)$ over the full range of τ. For short times, Eq. (34) gives $w(\tau)\propto(1-e^{-\beta\tau})^2=g_{ss}^{(2)}(\tau)$; it is still true that $w(0)=g_{ss}^{(2)}(0)=0$. Also, Fig. 1(a), and Eqs. (34) and (35), show a redistribution of waiting times qualitatively the same as Fig. 1(b): $w(\tau)<w_{\rm coh}(\tau)$, $\tau\ll\bar\tau$; $w(\tau)>w_{\rm coh}(\tau)$, $\tau\sim\bar\tau$; $w(\tau)<w_{\rm coh}(\tau)$, $\tau\gg\bar\tau$. However, the photoelectron counting sequences described by Eq. (34) differ from a random counting sequence by only a small amount. Only the rare photoelectric counts that follow a previous count by $\tau\lesssim 1/\beta\ll\bar\tau$ bring a difference from the exponential distribution of waiting times for coherent scattering. In this sense, the photon statistics in the weak-driving-field limit approach those for elastic dipole scattering, just as the fluorescent spectrum in the weak-driving-field limit approaches the spectrum for elastic scattering.

The nonclassical-character of photoelectron counting sequences also appears in the counting distribution $p_{ss}(n,T)\equiv\lim_{t\to\infty}p(n,t,t+T)$. The width of $p_{ss}(n,T)$ depends on the counting time T. The condition $g_{ss}^{(2)}(0)<1$ guarantees that $p_{ss}(n,T)$ will be sub-Poissonian in the short-counting-time limit ($\beta T\ll 1$).[4] Deviation from the Poisson condition $\Delta n^2=\bar n$ is measured by

$$\begin{aligned}Q(T)&=\overline{\Delta n^2}/\bar n-1\\&=\langle\hat I\rangle_{ss}T[g_{ss}^{(2)}(0)-1]\\&=-(\beta T)\frac{1}{2}\frac{\Omega^2}{\Omega^2+2\beta^2} .\end{aligned} \tag{37}$$

The deviation from a Poisson distribution is very small when $\beta T\ll 1$. For longer counting times larger deviations from Poisson counts are possible.[4,9] Figure 1(b) is plotted for the driving-field strength that maximizes $|Q|$ in the long-counting-time limit ($\beta T\gg 1$), with $Q(\infty)=-0.75$. The origin of the large sub-Poissonian effect is clear from the form of $w(\tau)$. If $w(\tau)=\delta(\tau-\bar\tau)$ there will be negligible variation in the number of photoelectric counts recorded in a time $T\gg\bar\tau$. The waiting-time distribution shown in Fig. 1(b) is highly peaked about $\bar\tau$. Therefore, reduced fluctuations in the number of photoelectric counts recorded for long counting times should be seen when waiting times obey this distribution rather than the exponential distribution for coherent light. This statement can be made quantitatively. Using Eq. (30), the variance $\Delta\tau^2$ is given by

$$\begin{aligned}\overline{\Delta\tau^2}&=\int_0^\infty d\tau\,\tau^2 w(\tau)-\bar\tau^2\\&=2\bar\tau^2\left[1-\frac{6\beta^2\Omega^2}{(\Omega^2+2\beta^2)^2}\right] .\end{aligned} \tag{38}$$

From Eq. (33),

$$\overline{\Delta\tau_{\rm coh}^2}=2\bar\tau^2 . \tag{39}$$

Thus,

$$\overline{\Delta\tau^2}/\overline{\Delta\tau_{\rm coh}^2}-1=-\frac{6\beta^2\Omega^2}{(\Omega^2+2\beta^2)^2} . \tag{40}$$

This is precisely the quantity $Q(\infty)$ characterizing the deviation of the photoelectron counting distribution from a Poissonian distribution for long counting times.[4]

In comparison with Fig. 1(b), Fig. 1(a) shows a very small peaking of $w(\tau)$ about $\bar\tau$. We therefore expect that $|Q(\infty)|\ll 1$ for the parameters of Fig. 1(a). This is what we find in Eq. (40); $|Q(\infty)|\ll 1$ when $\Omega^2/\beta^2\ll 1$. This comparison can be stated in terms of the two time scales that govern the form of $w(\tau)$: For $\Omega^2/\beta^2\lesssim 1$, a large sub-Poissonian effect in the long-counting-time limit requires the correlation time $1/\beta$ to be similar to the mean waiting time $\bar\tau$.

This criterion does not also hold in the strong-driving-field limit. In this limit the correlation time and the mean waiting time are the same, but Eq. (40) shows that $|Q(\infty)|\ll 1$. Figure 1(c) shows $w(\tau)$ for $\Omega^2/\beta^2\gg 1$. On the basis of the two curves shown here we can understand why fluctuations in the number of recorded counts in the long-counting-time limit should be similar for strongly driven resonance fluorescence and coherent scattering. If the Rabi oscillations are averaged in Fig. 1(c)—the first Rabi maximum against the second Rabi minimum, the second Rabi maximum against the third Rabi minimum, and so on—the average $\bar w(\tau)$ will rise from zero and then closely follow the exponential distribution $w_{\rm coh}(\tau)$—a form similar to that shown by Fig. 1(a). Then the departure from Poisson counts should be small by the ratio of the Rabi period $1/\Omega$ [the rise time for $\bar w(\tau)$] to the mean waiting time $\bar\tau\sim 1/\beta$ [the exponential decay time for $\bar w(\tau)$]. This is indeed what is found in Eq. (40) (variances actually give this ratio squared).

Nonunit detection efficiency

When considering nonunit detection efficiency a distinction must be made between photon emission sequences and sequences of recorded photoelectric counts. The formal relationship between these sequences is discussed in the next sections. Intuition tells us what to expect, however. When $\eta'\neq 1$, sequences of photon emissions are converted to sequences of photoelectrons by the random deletion of undetected photons (alternatively, the random selection of detected photons). As the rate of random deletion increases (selection decreases), the randomness of the detection process becomes increasingly important in determining the photoelectron statistics. When $\eta'\ll 1$, photoelectric counts are essentially recorded randomly, at a rate much lower than the rate at which

photons are emitted. The photoelectron waiting-time distribution approaches the exponential form for a random counting sequence.

The expression for $w(\tau)$ [Eq. (28)] does not have a simple form when $\eta' \neq 1$. However, setting $\Omega=\beta$ we find

$$w(\tau)=\tfrac{1}{3}\eta'\mu^{-2}\beta e^{-(1-\mu)\beta\tau} \times[1-2e^{-(3/2)\mu\beta\tau}\cos(\tfrac{1}{2}\sqrt{3}\mu\beta\tau-\pi/3)] , \quad (41)$$

where $\mu=(1-\eta')^{1/3}$. The average waiting time is

$$\bar{\tau}=(\eta\langle\hat{I}\rangle_{ss})^{-1} , \quad (42)$$

where

$$\eta\langle\hat{I}\rangle_{ss}=\eta'(2\beta)\frac{1}{2}\frac{\Omega^2}{\Omega^2+2\beta^2} . \quad (43)$$

For $\eta'=1$ Eq. (41) gives

$$w(\tau)=\tfrac{1}{2}\beta^3\tau^2 e^{-\beta\tau} , \quad (44)$$

and for $\eta' \ll 1$,

$$w(\tau)=\tfrac{1}{3}\eta'(1+\tfrac{2}{3}\eta')\beta\{\exp[-\tfrac{1}{3}\eta'(1+\tfrac{1}{3}\eta')\beta\tau] -2\exp(-\tfrac{3}{2}\beta\tau)\cos(\tfrac{1}{2}\sqrt{3}\beta\tau-\pi/3)\} . \quad (45)$$

In Eq. (45) we have kept terms in η' to sufficient order that $\int_0^\infty d\tau\, w(\tau)=1+O(\eta'^2)$.

Equation (45) shows the approach to an exponential distribution for low detection efficiency. It has a similar form to Eq. (34); $w(\tau)$ is less than $w_{\rm coh}(\tau)$ for short times, rises slightly above $w_{\rm coh}(\tau)$, and then falls below $w_{\rm coh}(\tau)$, decaying slightly faster than $w_{\rm coh}(\tau)$ for long times—in comparison with $w_{\rm coh}(\tau)$, $w(\tau)$ is *slightly* peaked about $\bar{\tau}$. The expected relationship [Eq. (20)] between $w(\tau)$ and $g_{ss}^{(2)}(\tau)$ for $\eta' \to 0$ also holds; we find

$$\lim_{\eta'\to 0}[w(\tau)/\tfrac{1}{3}\eta'\beta] =1-2e^{-(3/2)\beta\tau}\cos(\tfrac{1}{2}\sqrt{3}\beta\tau-\pi/3)=g_{ss}^{(2)}(\tau) .$$

The effect of reduced detection efficiency for other driving-field strengths is illustrated in Fig. 2. The same general features are seen. When $\bar{\tau}\propto 1/\eta'$ greatly exceeds the correlation time $1/\beta$, $w(\tau)$ approaches $w_{\rm coh}(\tau)$ for $\tau \gg 1/\beta$, while $w(\tau)/\eta\langle\hat{I}\rangle_{ss}$ approaches $g_{ss}^{(2)}(\tau)$ for $\tau \sim 1/\beta$.

III. CALCULATION OF EXCLUSIVE PROBABILITY DENSITIES FROM SOURCE DYNAMICS

In this section we begin the task of relating photoelectron counting statistics to the quantum dynamics of the atomic source. We work from Eq. (8) to show how the exclusive probability densities $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ can be evaluated directly in terms of dynamical equations for the source. The calculation is not difficult, but involves cumbersome notation. We therefore outline the approach for the simplest example. We first show how $w(\tau)$ can be calculated from dynamical equations for the source. The general expression for $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ can be inferred from this result.

The derivation of $w(\tau)$ from Eq. (21) relied on the evaluation of all the nonexclusive probability densities and their summation after taking Laplace transforms. The summation in Eq. (21) can also be performed in the time domain to give a compact time-domain expression for $w(\tau)$. We begin by displaying the time ordering and normal ordering explicitly. The photon flux operator is given by

$$\hat{I}_0(\tau)=\frac{2\epsilon_0 c}{\hbar\omega_A}\int_D dA\,\hat{\mathbf{E}}^{(-)}(\mathbf{r},t_0+\tau)\hat{\mathbf{E}}^{(+)}(\mathbf{r},t_0+\tau) , \quad (46)$$

where ω_A is the atomic resonance frequency and the integral is taken over the surface of the photoelectric detector. The electromagnetic field operators $\hat{\mathbf{E}}^{(\pm)}(\mathbf{r},t)$ may be written as the sum of free-field operators $\hat{\mathbf{E}}_f^{(\pm)}(\mathbf{r},t)$ and the source operators[21,22]

$$\hat{\mathbf{E}}_s^{(\pm)}(\mathbf{r},t)=\frac{\omega_A^2\mu\sin\theta}{4\pi\epsilon_0 c^2|\mathbf{r}-\mathbf{r}_A|}\mathbf{e}\hat{\sigma}_\mp(t-|\mathbf{r}-\mathbf{r}_A|/c) , \quad (47)$$

where $\hat{\sigma}_+=|+\rangle\langle-|$, and $\hat{\sigma}_-=|-\rangle\langle+|$ are atomic raising and lowering operators ($|+\rangle$ and $|-\rangle$ are the upper and lower states of the atomic transition), $\mathbf{r}_A$ is the position of the atom, $\boldsymbol{\mu}$ is the atomic dipole matrix element, θ is the angle between $\boldsymbol{\mu}$ and $\mathbf{r}-\mathbf{r}_A$, and $\mathbf{e}$ is a unit polarization vector (lying in the plane of $\boldsymbol{\mu}$ and $\mathbf{r}-\mathbf{r}_A$, perpendicular to $\mathbf{r}-\mathbf{r}_A$, with a positive projection on $\boldsymbol{\mu}$). Because of the explicit time and normal ordering, free-field operators can be dropped if the electromagnetic field is initially in the vacuum state.[23] We then write

$$\hat{I}_0(\tau)=\xi\mathcal{E}_s^{(-)}(t_0+\tau)\mathcal{E}_s^{(+)}(t_0+\tau) , \quad (48)$$

with collection efficiency $\xi=(8\pi/3)^{-1}\int_D d\Omega\sin^2\theta$, and

$$\mathcal{E}_s^{(\pm)}(t)=\sqrt{2\beta}\hat{\sigma}_\mp(t-r_{AD}/c) , \quad (49)$$

where r_{AD} is the distance from the atom to the detector.

Substituting from Eq. (48), and using the cyclic property of the trace, Eq. (21) can now be written in the form

$$\begin{aligned} w(\tau)=&\{\mathrm{tr}[\mathcal{E}_s^{(+)}(t_0)\mathcal{X}\mathcal{E}_s^{(-)}(t_0)]\}^{-1}\eta' \\ &\times\sum_{m=0}^{\infty}(-\eta')^m\int_0^\tau d\tau_m\int_0^{\tau_m}d\tau_{m-1}\cdots\int_0^{\tau_2}d\tau_1\mathrm{tr}[\,\mathcal{E}_s^{(+)}(t_0+\tau)\mathcal{E}_s^{(+)}(t_0+\tau_m)\mathcal{E}_s^{(+)}(t_0+\tau_{m-1})\cdots\mathcal{E}_s^{(+)}(t_0+\tau_1) \\ &\qquad\times\mathcal{E}_s^{(+)}(t_0)\mathcal{X}\mathcal{E}_s^{(-)}(t_0)\mathcal{E}_s^{(-)}(t_0+\tau_1)\cdots\mathcal{E}_s^{(-)}(t_0+\tau_{m-1}) \\ &\qquad\times\mathcal{E}_s^{(-)}(t_0+\tau_m)\mathcal{E}_s^{(-)}(t_0+\tau)] \\ =&[\mathrm{tr}(\mathcal{S}\mathcal{X}_0)]^{-1}\eta'\sum_{m=0}^{\infty}(-\eta')^m\int_0^\tau d\tau_m\int_0^{\tau_m}d\tau_{m-1}\cdots\int_0^{\tau_2}d\tau_1\mathrm{tr}[\mathcal{S}e^{L(\tau-\tau_m)}\mathcal{S}e^{L(\tau_m-\tau_{m-1})}\cdots e^{(L\tau_1)}\mathcal{S}\mathcal{X}_0] , \end{aligned} \quad (50)$$

with

$$\hat{\chi}_0 \equiv \hat{\chi}(t_0 - r_{AD}/c) = \exp[L(t_0 - r_{AD}/c)]\hat{\chi} \ . \tag{51}$$

$\hat{\chi}$ is the initial density operator for the electromagnetic field and its source, and L and $\mathcal{S}$ operate on the space of Hilbert space operators; for any operator $\hat{O}$,

$$L\hat{O} = (1/i\hbar)[\hat{H}, \hat{O}] \ , \tag{52}$$

$$\mathcal{S}\hat{O} = \mathcal{E}_s^{(+)}(r_{AD}/c)\hat{O}\mathcal{E}_s^{(-)}(r_{AD}/c) = (2\beta)\hat{\sigma}_- \hat{O} \hat{\sigma}_+ \ , \tag{53}$$

where $\hat{H}$ is the Hamiltonian in a frame rotating at the frequency ω_A (we assume on resonance excitation):

$$\hat{H} = \sum_{\mathbf{k},\lambda} \hbar(\omega_k - \omega_A)\hat{r}^{\dagger}_{\mathbf{k},\lambda}\hat{r}_{\mathbf{k},\lambda} + \sum_{\mathbf{k},\lambda} \hbar(\kappa_{\mathbf{k},\lambda}\hat{r}_{\mathbf{k},\lambda}\hat{\sigma}_+ + \kappa^*_{\mathbf{k},\lambda}\hat{r}^{\dagger}_{\mathbf{k},\lambda}\hat{\sigma}_-) - \hbar(\Omega/2)(\hat{\sigma}_+ + \hat{\sigma}_-) \ , \tag{54}$$

where $\hat{r}^{\dagger}_{\mathbf{k},\lambda}$ and $\hat{r}_{\mathbf{k},\lambda}$ are creation and annihilation operators for the electromagnetic field mode with frequency ω_k, wave vector $\mathbf{k}$, and polarization state λ, $\kappa_{\mathbf{k},\lambda}$ is a coupling constant, and Ω is the Rabi frequency for the driving field. The sum in Eq. (50) is evaluated using the identity

$$\exp[(L + a\mathcal{S})\tau] = \sum_{m=0}^{\infty} a^m \int_0^{\tau} d\tau_m \int_0^{\tau_m} d\tau_{m-1} \cdots \int_0^{\tau_2} d\tau_1 \exp[L(\tau - \tau_m)]\mathcal{S}\exp[L(\tau_m - \tau_{m-1})]\mathcal{S}\cdots\mathcal{S}\exp(L\tau_1) \ . \tag{55}$$

Then

$$w(\tau) = [\mathrm{tr}(\mathcal{S}\hat{\chi}_0)]^{-1}\eta' \,\mathrm{tr}[\mathcal{S}e^{(L-\eta'\mathcal{S})\tau}\mathcal{S}\hat{\chi}_0] \ . \tag{56}$$

Equation (56) gives a simple formal expression for the waiting-time distribution; but it is not very useful for an explicit evaluation of $w(\tau)$. The difficulty is that L acts in the full space of the electromagnetic field and the source, and the trace is to be taken over the many modes of the electromagnetic filed as well as the source. We can remove this difficulty by adopting the methods used in a master equation treatment of source dynamics. Using these methods we can perform the trace over the electromagnetic field *a priori,* leaving an expression to be evaluated in the space of the source alone. The master equation for single-atom resonance fluorescence reads[1]

$$\dot{\hat{\rho}} = \mathcal{L}\hat{\rho} \ , \tag{57}$$

with

$$\mathcal{L}\hat{\rho} = i(\Omega/2)[\hat{\sigma}_+ + \hat{\sigma}_-, \hat{\rho}] + \beta(2\hat{\sigma}_-\hat{\rho}\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_-\hat{\rho} - \hat{\rho}\hat{\sigma}_+\hat{\sigma}_-) \ , \tag{58}$$

where the source density operator $\hat{\rho}$ is defined by

$$\hat{\rho} = \mathrm{tr}_{\mathrm{field}}[\hat{\chi}(t)] \ . \tag{59}$$

The trace over field modes in Eq. (56) is evaluated in the same approximation used to derive Eq. (57) from the Liouville equation $\dot{\hat{\chi}} = L\hat{\chi}$. This simply results in the replacements $L \to \mathcal{L}$ and $\hat{\chi} \to \hat{\rho}$. We have

$$w(\tau) = [\mathrm{tr}(\mathcal{S}\hat{\rho}_0)]^{-1}\eta' \mathrm{tr}[\mathcal{S}\exp[(\mathcal{L} - \eta'\mathcal{S})\tau]\mathcal{S}\hat{\rho}_0] \ . \tag{60}$$

The expression on the right-hand side of Eq. (60) may be evaluated by solving a set of modified Bloch equations for the driven atom.

Before we describe the formulation in terms of modified Bloch equations, let us say something about the formal structure of Eq. (60); this structure has a simple and instructive physical interpretation. The waiting-time distribution is defined by the probability density

$$\wp_1(t_0 + \tau | t_0) \equiv \mathfrak{p}_a(\tau) = \mathfrak{p}_g(\tau)p_0(\tau) \ , \tag{61}$$

where $\mathfrak{p}_a(\tau)$ is the probability density for recording a photoelectric count at $t_0 + \tau$ *and* no counts in the interval t_0 to $t_0 + \tau$, given a photoelectric count is recorded at t_0; $\mathfrak{p}_g(\tau)$ is the probability density for recording a photoelectric count at $t_0 + \tau$, *given* no counts are recorded in the interval t_0 to $t_0 + \tau$ given a photoelectric count is recorded at t_0; and $p_0(\tau)$ is the probability that no counts are recorded in the interval t_0 to $t_0 + \tau$ given a photoelectric count is recorded at t_0. Equation (60) can be written as such a product:

$$w(\tau) = \eta' \mathrm{tr}[\mathcal{S}\hat{\bar{\rho}}_{\eta'}(\tau)]\mathrm{tr}\{\exp[(\mathcal{L} - \eta'\mathcal{S})\tau]\hat{\bar{\rho}}_0\} \ , \tag{62}$$

where

$$\hat{\bar{\rho}}_{\eta'}(\tau) = \frac{\exp[(\mathcal{L} - \eta'\mathcal{S})\tau]\hat{\bar{\rho}}_0}{\mathrm{tr}\{\exp(\mathcal{L} - \eta'\mathcal{S})\tau]\hat{\bar{\rho}}_0\}} \tag{63}$$

is a source density operator, with

$$\hat{\bar{\rho}}_{\eta'}(0) = \hat{\bar{\rho}}_0 = \frac{\mathcal{S}\hat{\rho}_0}{\mathrm{tr}(\mathcal{S}\hat{\rho}_0)} = \frac{\mathcal{S}\hat{\rho}(t_0 - r_{AD}/c)}{\mathrm{tr}[\mathcal{S}\hat{\rho}(t_0 - r_{AD}/c)]} \ ; \tag{64}$$

using Eq. (53)

$$\hat{\bar{\rho}}_{\eta'}(0) = \hat{\bar{\rho}}_0 = |-\rangle\langle -| \ . \tag{65}$$

From Eqs. (61) and (62) we identity

$$\mathfrak{p}_g(\tau) = \eta' \mathrm{tr}[\mathcal{S}\hat{\bar{\rho}}_{\eta'}(\tau)] = \eta'(2\beta)\langle + |\hat{\bar{\rho}}_{\eta'}(\tau)| + \rangle \ , \tag{66}$$

and

$$p_0(\tau) = \mathrm{tr}\{\exp[(\mathcal{L} - \eta'\mathcal{S})\tau]\hat{\bar{\rho}}_0\} = \mathrm{tr}\{\exp[(\mathcal{L} - \eta'\mathcal{S})\tau]|-\rangle\langle -|\} \ . \tag{67}$$

The physical significance of the density operator $\hat{\bar{\rho}}_{\eta'}(\tau)$ is

clear from Eq. (66). The right-hand side of this equation is just the product of the detection probability η' and the photon flux from the source, calculated with respect to $\hat{\hat{\rho}}_{\eta'}(\tau)$. Thus, $\hat{\hat{\rho}}_{\eta'}(\tau)$ describes the reduced state of the source (at the retarded time $t_0+\tau-r_{AD}/c$) inferred from the measurement, with efficiency η', recording a photoelectric count at t_0 and no counts in the interval t_0 to $t_0+\tau$. The density operator $\hat{\hat{\rho}}_{\eta'}=\hat{\hat{\rho}}_0$ describes the reduced state of the source inferred from the photocount recorded at t_0 alone; as expected, this count tells us that the atom was in the ground state at t_0-r_{AD}/c.

To actually evaluate the expression on the right-hand side of Eq. (62) we write this equation in the equivalent form

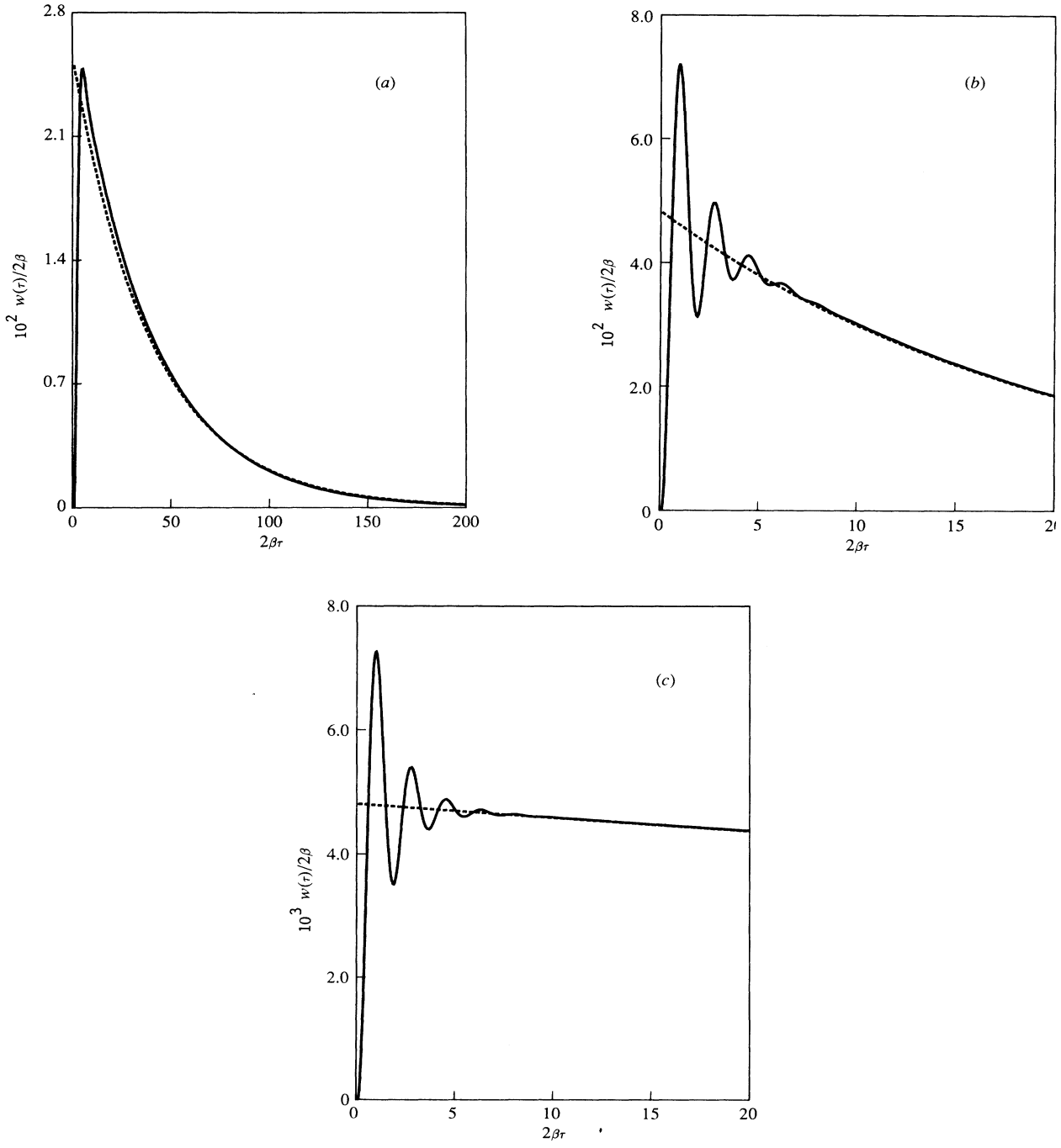

FIG. 2. Comparison of the photoelectron waiting-time distributions for resonance fluorescence and coherent light of equal intensity: (a) $\Omega/\sqrt{2}\beta=1$, $\eta'=0.1$; (b) $\Omega/\sqrt{2}\beta=5$, $\eta'=0.1$; and (c) $\Omega/\sqrt{2}\beta=5$, $\eta'=0.01$.

$$w(\tau)=\eta' \mathrm{tr}\{\mathcal{S}\exp[(\mathcal{L}-\eta'\mathcal{S})\tau]\hat{\rho}_0\}$$
$$=\eta'(2\beta)\langle +|\{\exp[(\mathcal{L}-\eta'\mathcal{S})\tau]|-\rangle\langle -|\}|+\rangle \ . \tag{68}$$

The action of the propagator $\exp[(\mathcal{L}-\eta'\mathcal{S})\tau]$ on an operator $\hat{O}$ is found by solving the equation

$$\dot{\hat{O}}=(\mathcal{L}-\eta'\mathcal{S})\hat{O} \ , \tag{69}$$

where

$$(\mathcal{L}-\eta'\mathcal{S})\hat{O}=i(\Omega/2)[\hat{\sigma}_+ +\hat{\sigma}_-,\hat{\rho}]+(1-\eta')(2\beta)\hat{\sigma}_-\hat{O}\hat{\sigma}_+$$
$$-\beta(\hat{\sigma}_+\hat{\sigma}_-\hat{O}+\hat{O}\hat{\sigma}_+\hat{\sigma}_-) \ . \tag{70}$$

[Note that Eq. (69) does not conserve $\mathrm{tr}(\hat{O})$, which is why the explicit normalization appears in Eq. (63).] Written in matrix element form, Eq. (69) reads

$$\dot{\mathbf{O}}=M\mathbf{O} \tag{71}$$

with

$$M=\begin{bmatrix} -2\beta & 0 & 0 & \Omega \\ (1-\eta')2\beta & 0 & 0 & -\Omega \\ 0 & 0 & -\beta & 0 \\ -\Omega/2 & \Omega/2 & 0 & -\beta \end{bmatrix} , \tag{72}$$

where $\mathbf{O}$ has the vector components

$$O_1=\langle +|\hat{O}|+\rangle, \quad O_2=\langle -|\hat{O}|-\rangle \ ,$$
$$O_3=\mathrm{Re}(\langle +|\hat{O}|-\rangle) \ , \tag{73}$$
$$O_4=\mathrm{Im}(\langle +|\hat{O}|-\rangle) \ .$$

When $\eta'=0$, Eq. (71) reduces to the normal Bloch equations.

The waiting-time distribution is obtained by solving Eq. (71) with initial conditions $O_1(0)=O_3(0)=O_4(0)=0$, $O_2(0)=1$. Then $w(\tau)=\eta'(2\beta)O_1(\tau)$. This calculation reproduces the result given in Eq. (28). The reduced state of the source inferred during the waiting times between photoelectric counts is given by

$$\hat{\bar{\rho}}_{\eta'}(\tau)=\frac{O_1(\tau)|+\rangle\langle +|+O_2(\tau)|-\rangle\langle -|+iO_4(\tau)(|+\rangle\langle -|-|-\rangle\langle +|)}{O_1(\tau)+O_2(\tau)} \ . \tag{74}$$

There is no dependence on $O_3(\tau)$ since the equation for O_3 decouples from the remaining three equations and $O_3(\tau)=\exp(-\beta\tau)O_3(0)=0$.

A generalization of the argument leading to Eq. (60) gives an expression for the exclusive probability densities $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ in terms of dynamical equations for the source (an alternative derivation appears in Appendix A of Ref. 23). We find

$$\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$$
$$=\eta'^{\,m}\mathrm{tr}\{\exp[(\mathcal{L}-\eta'\mathcal{S})(t+T-t_m)]\mathcal{S}\cdots\mathcal{S}\exp[(\mathcal{L}-\eta'\mathcal{S})(t_2-t_1)]\mathcal{S}\exp[(\mathcal{L}-\eta'\mathcal{S})(t_1-t)]\hat{\rho}(t-r_{AD}/c)\} \ . \tag{75}$$

Equation (60) follows as a special case of this result, when, from Eqs. (2) and (4), we write $w(\tau)=\wp_2(t_0,t_0+\tau;[t_0,t_0+\tau])/w_1(t_0)$. Using the specific form of $\mathcal{S}$ defined in Eq. (53), Eq. (75) factorizes as

$$\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])=\{\wp_1(t_m;[t_m,t+T])/w_1(t_m)\}\left[\sum_{i=2}^{m} w(t_i-t_{i-1})\right]\wp_1(t_1;[t,t_1]) \ , \tag{76}$$

where $w(t_i-t_{i-1})$ is given by Eq. (68),

$$\wp_1(t_m;[t_m,t+T])/w_1(t_m)$$
$$=\mathrm{tr}\{\exp[(\mathcal{L}-\eta'\mathcal{S})(t+T-t_m)]|-\rangle\langle -|\} \tag{77}$$

is the probability for no photoelectric counts in the interval t_m to $t+T$ given a photoelectric count is recorded at t_m, and

$$\wp_1(t_1;[t,t_1])=\eta'(2\beta)\langle +|\{\exp[(\mathcal{L}-\eta'\mathcal{S})(t_1-t)]$$
$$\times\hat{\rho}(t-r_{AD}/c)\}|+\rangle \tag{78}$$

is the probability density that the first photoelectric count is recorded at t_1 if observation begins at t. Equation (76) establishes the Markov and stationary (after the first count) character of photoelectric counting sequences for single-atom resonance fluorescence.

IV. DERIVATION OF THE PHOTOELECTRON COUNTING DISTRIBUTION FROM EXCLUSIVE PROBABILITY DENSITIES

We have derived Eq. (75) from the theory of photoelectric detection, which describes the sequences of photoelectrons generated by a field illuminating the photocathode of a photoelectric detector. The connection with source dynamics was made using Eq. (47) to express the field at the detector in terms of the dipole operators of the radiating atom. It is also possible to arrive at Eq. (75) without referring to the theory of photoelectric detection at all, using the master equation for the atom to follow

the sequences of photon emissions. We will describe this alternative approach in the next section, but first let us calculate the photoelectron counting distribution from our result for the exclusive probability densities.

The photoelectron counting distribution for single-atom resonance fluorescence has been calculated by a number of authors. Most of these calculations[4,7–10] use the Kelly-Kleiner formula[17]

$$p(n,t,t+T)=\left\langle \mathcal{T}:\frac{[\hat{\Omega}(t+T,t)]^n}{n!}\exp[-\hat{\Omega}(t+T,t)]:\right\rangle , \tag{79}$$

where $\hat{\Omega}(t+T,t)$ is defined by Eq. (9). The right-hand side of Eq. (79) can be evaluated in a series of steps parallel to the calculation of $w(\tau)$ in Sec. II. The exponential is expanded to give

$$p(n,t,t+T)=\eta^n\sum_{m=0}^{\infty}(-\eta)^m\frac{(n+m)!}{n!m!}\int_0^T d\tau_{n+m}\times\int_0^{\tau_{n+m}}d\tau_{n+m-1}\cdots\int_0^{\tau_2}d\tau_1\langle \mathcal{T}:\hat{I}(t+\tau_{n+m})\times\hat{I}(t+\tau_{n+m-1})\cdots\hat{I}(t+\tau_1):\rangle , \tag{80}$$

then the correlation functions in the integrand factorize as in Eqs. (22) and (23), and the summation can be performed after taking Laplace transforms with respect to T. The calculation of $p(n,t,t+T)$ from exclusive probability densities is a little more straightforward. From Eqs. (5) and (76) we have

$$p(n,t,t+T)=\int_0^T d\tau_n\int_0^{\tau_n}d\tau_{n-1}\cdots\int_0^{\tau_2}d\tau_1 p_0(T-\tau_n)w(\tau_n-\tau_{n-1})\cdots w(\tau_2-\tau_1)p_1(\tau_1|t) , \tag{81}$$

where $\tau_m=t_m-t$, and we have defined [Eqs. (77) and (78)]

$$p_0(T-\tau_n)\equiv\wp_1(t+\tau_n;[t+\tau_n,t+T])/w_1(t+\tau_n)=\mathrm{tr}\{\exp[(\mathcal{L}-\eta'\mathcal{S})(T-\tau_n)]|-\rangle\langle -|\} , \tag{82}$$

and

$$p_1(\tau_1|t)\equiv\wp_1(t+\tau_1;[t,t+\tau_1])=\eta'(2\beta)\langle +|\exp[(\mathcal{L}-\eta'\mathcal{S})\tau_1]\hat{\rho}(t-r_{AD}/c)|+\rangle . \tag{83}$$

Taking Laplace transforms, Eq. (81) gives

$$\tilde{p}(n,s,t)=\tilde{p}_0(s)[\tilde{w}(s)]^{n-1}\tilde{p}_1(s|t) . \tag{84}$$

$\tilde{p}(n,s,t)$ is the Laplace transform of $p(n,t,t+T)$ with respect to T, and $\tilde{p}_0(s)$ and $\tilde{p}_1(s|t)$ are the Laplace transforms, with respect to τ, of $p_0(\tau)$ and $p_1(\tau|t)$, respectively. Equation (84) displays the underlying structure of the sequence of photoelectric counts—a first count described by $p_1(\tau|t)$, followed by $n-1$ counts described by $w(\tau_i-\tau_{i-1})$, $i=2,\ldots,n$, and finally an interval without counts described by $p_0(T-\tau_n)$. Explicit expressions for $\tilde{w}(s)$ and $\tilde{p}_0(s)$ are found by solving Eq. (71) with $\hat{O}(0)=|-\rangle\langle -|$, and the explicit expression for $\tilde{p}_1(s|t)$ is found by solving the same equation with $\hat{O}(0)=\hat{\rho}(t-r_{AD}/c)$. Equation (27) gives the result for $\tilde{w}(s)$. For $\tilde{p}_0(s)$ and $\tilde{p}_1(\tau|t)$ we find

$$\tilde{p}_0(s)=\frac{(s+\beta)(s+2\beta)+\Omega^2}{s(s+\beta)(s+2\beta)+\Omega^2(s+\eta'\beta)} , \tag{85}$$

and

$$\tilde{p}_1(s|t)=\eta'(2\beta)\frac{s(s+\beta)\langle +|\hat{\rho}_t|+\rangle-s\Omega\,\mathrm{Im}\langle +|\hat{\rho}_t|-\rangle+\Omega^2/2}{s(s+\beta)(s+2\beta)+\Omega^2(s+\eta'\beta)} , \tag{86}$$

where $\hat{\rho}_t=\hat{\rho}(t-r_{AD}/c)$.

Of course, the simple structure of Eq. (84) is lost in the time domain. Indeed, inversion of the Laplace transform leads to a very complicated expression for $p(n,t,t+T)$. Actually, the moments of $p(n,t,t+T)$ are more relevant to experimental measurements than the full counting distribution. Low-order moments are given by relatively simple expressions, and, in particular, much attention has been paid to the quantity $Q=\Delta n^2/\bar{n}-1$ which characterizes the sub-Poissonian character of the photoelectric counts.[4,9,10] Explicit results for $p(n,t,t+T)$ are still useful, however, for illustrating the sub-Poissonian statistics. Since very few plots of the counting distribution have appeared in the literature we present a number here.

Most of the published expressions for $p(n,t,t+T)$ are limited to special cases, such as unit detection efficiency,[4–6,8] short or long counting times,[4,7,8] or weak or strong driving fields.[5,6] Singh has given a general analytic result for resonant excitation.[9] A slightly different algorithm was used to generate the curves plotted in Figs. 3 and 4. This algorithm is described in the Appendix. Figures 3 and 4 compare the photoelectron counting distribution for single-atom resonance fluorescence with that for coherent scattering of the same intensity. Both

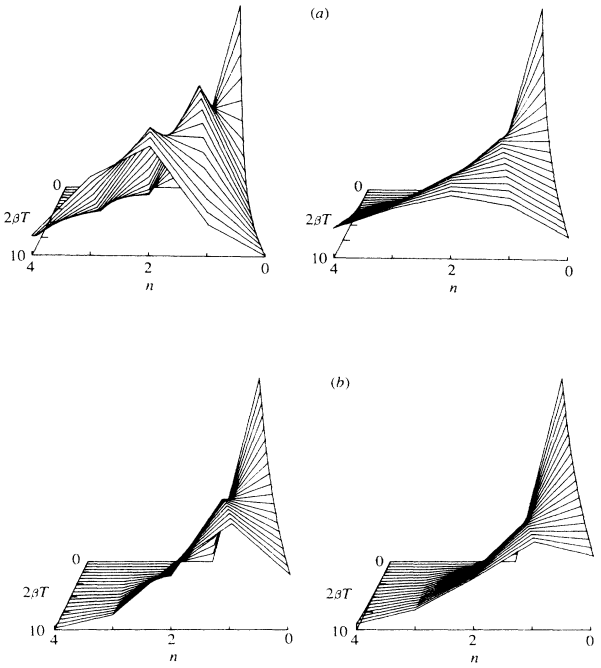

FIG. 3. Comparison of the photoelectron counting distributions for resonance fluorescence (on the left) and coherent light of equal intensity (on the right) for short counting times T: $\Omega/\sqrt{2}\beta=1$, and (a) $\eta'=1$, (b) $\eta'=0.5$.

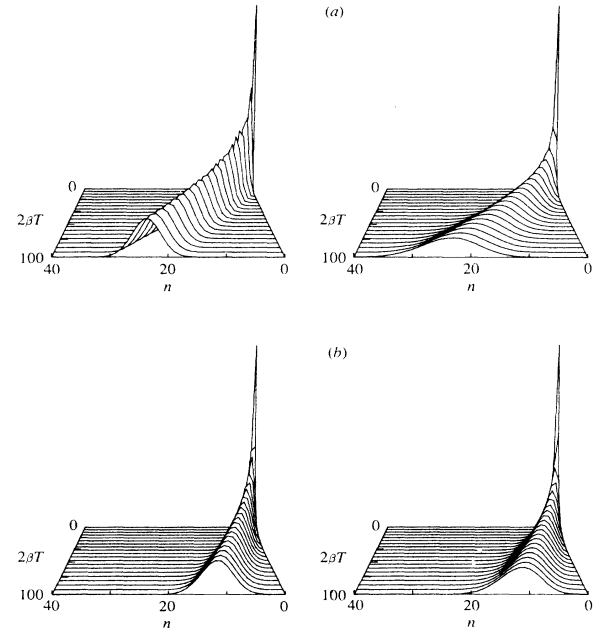

FIG. 4. Comparison of the photoelectron counting distributions for resonance fluorescence (on the left) and coherent light of equal intensity (on the right) for long counting times T: $\Omega/\sqrt{2}\beta=1$, and (a) $\eta'=1$, (b) $\eta'=0.5$.

figures are plotted for the steady-state limit and $\Omega=\sqrt{2}\beta$, conditions that give the largest sub-Poissonian effect, with $Q=-0.75$ when $\eta'=1$. For this value of Q, $p(n,t,t+T)$ should have half the width of the counting distribution for coherent scattering. This is indeed what we find in Fig. 4(a). The reduced width is due to the reduced variance of the waiting-time distribution illustrated in Fig. 1(b). The degradation of sub-Poissonian statistics by a nonunit detection efficiency is clearly illustrated by the comparison between Figs. 3(a) and 3(b), and 4(a) and 4(b), and correlates with the broadening of the waiting-time distribution illustrated in Fig. 2(a).

V. SOURCE DYNAMICS AND ATOMIC STATE REDUCTION

The derivations of the photoelectron counting distribution by Mollow[5] and Cook[6] differ from those of other authors by explicitly counting photon emissions rather than photoelectrons. Mollow's work is based on a hierarchy of equations that give the probabilities for finding n photons in the many modes of the fluorescent field. These equations are used by Zoller *et al.*[12] in their analysis of quantum jumps. Cook derives the probability for n photon emissions from the hierarchy of equations describing momentum transfer from the driving field to the atom, counting the number of momentum "kicks" imparted to the atom. Mollow and Cook formulate their calculations quite differently, but the hierarchies of equations which they use are actually equivalent. Both effectively calculate the photoelectron counting distribution for $\eta'=1$ from exclusive probability densities. The connection is made clear in this section, where we complete our demonstration of the formal relationship between photoelectron counting sequences and photon emission sequences. We derive the expression for exclusive probability densities given in Eq. (75) from the master equation for the source, without reference to the theory of photoelectric detection. We first calculate the probability densities $\wp_m^e(t_1,t_2,\ldots,t_m;[t,t+T])$ for m photon emissions in the interval $[t-r_{AD}/c,t+T-r_{AD}/c]$ at times t_1-r_{AD}/c, $t_2-r_{AD}/c,\ldots,t_m-r_{AD}/c$. Our derivation, although more formal, follows an approach equivalent to that taken by Mollow and Cook. We then show how a nonunit detection efficiency can be included to arrive at Eq. (75).

We have a second goal in this section. That is to clarify the issue of atomic state reduction. Cohen-Tannoudji first raised this issue in his Les Houche lectures in 1975.[24] His discussion of photon antibunching includes the comment, "The *detection* of this first photon 'reduces the wave packet.' Immediately after this detection the atom is certainly in its lower state," and the questions, "Is it possible to consider such an experiment as a possible test of the postulate of the reduction of the wave packet? What would be the predictions of other quantum theories of measurement?" The italics are our own; according to Cohen-Tannoudji's comment the *detection* of a photon reduces the wave packet. The implication is that photoelectric detection in some sense *causes* the reduction of the wave packet. Pegg states this viewpoint explicitly in

his treatment of photon antibunching by absorber theory: "In absorber theory an upward transition in the detector atom c, which constitutes the measurement process, can be interpreted as the actual physical *cause* of the corresponding reversion of the atom to its ground state despite the fact that the transition in c occurs *after* a has reverted to its ground state."[25] The idea that photoelectric detection plays a causative role in atomic state reduction appears again in recent work on quantum jumps: "The *detection* of this photon then projects the atom in a lower manifold;"[26] ". . . giving rise to discontinuities or jumps *associated with the detection process*;"[27] "These experiments have a special fascination, . . . , they allow one to watch the reduction of the wave function *by the measurement process* on the oscilloscope screen."[28] (emphases added).

What role does photoelectric detection actually play in the return of the atom to its ground state after each photon emission? Indeed, what does it mean to speak of photon emissions as realized events separate from photoelectron detection? We offer an answer to these questions based on the calculation that follows. The viewpoint for which we argue holds that photoelectric detection does not cause atomic state reduction. Projection of the atom into its ground state is caused by the *dissipative* nature of the atomic dynamics, and reoccurs, on average, at the mean spontaneous emission rate, with complete indifference to the presence or absence of an observer. Photoelectric detection merely monitors emitted (realized) photons. It does not intrude into a coherent quantum dynamic in the manner implied by a measurement-induced wave packet reduction; it is the *irreversible* decay into the vacuum that interrupts the coherence of the source dynamics. No doubt, in general outline, this viewpoint is already widely held; it is the natural one suggested by intuition. However, it is rarely recognized explicitly and has not been stated succinctly in the theoretical literature on resonance fluorescence.

We begin with the master equation for the atom plus quantized driving field:

$$\dot{\hat{\rho}}' = \mathcal{L}'\hat{\rho}' , \tag{87}$$

with

$$\mathcal{L}'\hat{\rho}' = ig[\hat{a}\hat{\sigma}_+ + a^\dagger\hat{\sigma}_-, \hat{\rho}'] + \beta(2\hat{\sigma}_-\rho'\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_-\rho' - \rho'\hat{\sigma}_+\hat{\sigma}_-) , \tag{88}$$

where

$$g = \left[\frac{\omega_A \mu^2}{2\hbar\epsilon_0 V}\right]^{1/2} \tag{89}$$

is the atom-field coupling constant, and $\hat{\rho}'$ is the density operator for the coupled atom-field source. We used a classical driving field in Sec. III [Eq. (58)]. Quantizing the driving field now is simply a temporary notational device that allows us to track photon emissions using the quantum number on the energy states of the source. The number of photons emitted during any time interval is given by the number of energy quanta lost during that interval from the source. Thus, energy quanta lost by the source can be used for the same purpose that Cook uses the momentum quanta gained by the atom. We will eventually recover results for a classical driving field by taking the formal limit of infinite photon number and finite energy density. If the initial state of the driving field has mean photon number n_0 and photon number variance σ^2, this limit is taken with

$$\begin{aligned} &n_0 \to \infty, \quad g \to 0 \ (V \to \infty) , \\ &\sigma/n_0 \to 0, \quad g\sqrt{n_0} = \Omega/2 \text{ const} . \end{aligned} \tag{90}$$

In this limit

$$\mathrm{tr}_f(\mathcal{L}'\hat{\rho}') = \mathcal{L}(\mathrm{tr}_f\hat{\rho}') = \mathcal{L}\hat{\rho} , \tag{91}$$

where the trace is taken over the driving field; Eq. (87) then reduces to Eq. (57).

The formal solution to Eq. (87) has

$$\hat{\rho}'(t + T - r_{AD}/c) = \exp(\mathcal{L}'T)\hat{\rho}'(t - r_{AD}/c) . \tag{92}$$

We wish to separate the time evolution generated by $\exp(\mathcal{L}'T)$ into two parts: evolution that is confined to each manifold of source states with a definite number of energy quanta, and evolution between these manifolds, accompanied by the loss of one energy quantum from the source. We first introduce a projection operator $\mathcal{P}$ that separates the (block) diagonal part of the density operator describing source states with definite numbers of energy quanta:

$$\mathcal{P}\hat{\rho}' = \sum_{n=0}^{\infty} \hat{\rho}'_n , \tag{93}$$

where

$$\hat{\rho}'_0 = |0,-\rangle\langle 0,-|\rho'_{0,-;0,-} , \tag{94}$$

and, for $n \geq 1$,

$$\hat{\rho}'_n = (|n-1,+\rangle |n,-\rangle) \begin{bmatrix} \rho'_{n-1,+;n-1,+} & \rho'_{n-1,+;n,-} \\ \rho'_{n,-;n-1,+} & \rho'_{n,-;n,-} \end{bmatrix} \times \begin{bmatrix} \langle n-1,+| \\ \langle n,-| \end{bmatrix} . \tag{95}$$

It can be shown that $\mathcal{P}\hat{\rho}'$ evolves independently of $(1-\mathcal{P})\hat{\rho}'$; formally, that $\mathcal{P}$ commutes with $\mathcal{L}'$. Thus,

$$\mathcal{P}\hat{\rho}'(t + T - r_{AD}/c) = \exp(\mathcal{L}'T)\mathcal{P}\hat{\rho}'(t - r_{AD}/c) . \tag{96}$$

We now decompose the source dynamics by writing

$$\mathcal{L}' = (\mathcal{L}' - \mathcal{S}) + \mathcal{S} . \tag{97}$$

$\mathcal{L}' - \mathcal{S}$ has the property that it only couples matrix elements within the manifold spanned by $|n-1,+\rangle$ and $|n,-\rangle$ to other matrix elements within that manifold [also $(\mathcal{L}' - \mathcal{S})\hat{\rho}'_0 = 0$]. $\mathcal{S}$ couples the manifold spanned by states with n energy quanta to the manifold spanned by states with $n-1$ energy quanta:

$$\mathcal{S}\hat{\rho}'_n = \rho'_{n-1,+;n-1,+}|n-1,-\rangle\langle n-1,-| . \tag{98}$$

Note that $\mathcal{S}$ also sets the atom in its ground state, accomplishing the atomic-state reduction seen in Eqs. (64) and (65). Using Eq. (96) and the identity (55) we now write

$$\mathcal{P}\hat{\rho}'(t+T-r_{AD}/c)=\exp\{[(\mathcal{L}'-\mathcal{S})+\mathcal{S}]T\}\mathcal{P}\hat{\rho}'(t-r_{AD}/c)$$

$$=\sum_{m=0}^{\infty}\int_t^{t+T}dt_m\int_t^{t_m}dt_{m-1}\cdots\int_t^{t_2}dt_1\exp[(\mathcal{L}'-\mathcal{S})(t+T-t_m)]\mathcal{S}$$

$$\times\exp[(\mathcal{L}'-\mathcal{S})(t_m-t_{m-1})]\mathcal{S}\cdots\mathcal{S}$$

$$\times\exp[(\mathcal{L}'-\mathcal{S})(t_1-t)]\mathcal{P}\hat{\rho}'(t-r_{AD}/c)\ . \tag{99}$$

The exercise we have performed here merely isolates the part of the source dynamics associated with energy loss into the fluorescent field. Use of the projector $\mathcal{P}$ is not, in fact, essential. We could write Eq. (99) with $\mathcal{P}\hat{\rho}'$ replaced by $\hat{\rho}'$, as a decomposition of Eq. (92). However, the projector allows us to visualize the significance of $\mathcal{S}$ and $\mathcal{L}'-\mathcal{S}$ more readily. $\mathcal{P}\hat{\rho}'(t-r_{AD}/c)$ is a density operator describing a statistical mixture of source states with definite numbers of energy quanta at time $t-r_{AD}/c$:

$$\mathcal{P}\hat{\rho}'(t-r_{AD}/c)=\sum_{n=0}^{\infty}p_n(t-r_{AD}/c)\frac{\hat{\rho}'_n(t-r_{AD}/c)}{\mathrm{tr}[\hat{\rho}'_n(t-r_{AD}/c)]}\ , \tag{100}$$

where $p_n=\mathrm{tr}(\hat{\rho}'_n)=\rho'_{n-1,+;n-1,+}+\rho'_{n,-;n,-}$ is the probability that the source contains n energy quanta. Under the action of $\mathcal{S}$ and $\mathcal{L}'-\mathcal{S}$ in the integrand of Eq. (99), a source state with n energy quanta at time $t-r_{AD}/c$, evolves into a state with $n-m$ quanta at time $t+T-r_{AD}/c$. The photon emissions are generated by the m appearances of $\mathcal{S}$, separated by intervals of evolution without photon emission generated by $\mathcal{L}'-\mathcal{S}$. If we define the probability for n photon emissions during the interval $[t-r_{AD}/c,t+T-r_{AD}/c]$ by

$$p_e(n,t,t+T)\equiv\mathfrak{p}_n(t,t+T) \tag{101}$$

where $\mathfrak{p}_n(t,t+T)$ is the probability that the source is in a state at $t+T-r_{AD}/c$ reached from $\hat{\rho}'(t-r_{AD}|c)$ with the loss of n energy quanta, we can write

$$p_e(n,t,t+T)=\int_t^{t+T}dt_m\int_t^{t_m}dt_{m-1}\cdots\int_t^{t_2}dt_1\wp_m^e(t_1,t_2,\ldots,t_m;[t,t+T])\ , \tag{102}$$

where

$$\wp_m^e(t_1,t_2,\ldots,t_m;[t,t+T])$$
$$=\mathrm{tr}\{\exp[(\mathcal{L}'-\mathcal{S})(t+T-t_m)]\mathcal{S}\cdots\mathcal{S}\exp[(\mathcal{L}'-\mathcal{S})(t_2-t_1)]\mathcal{S}\exp[(\mathcal{L}'-\mathcal{S})(t_1-t)]\mathcal{P}\hat{\rho}'(t-r_{AD}/c)\}\ . \tag{103}$$

In the limit defined by Eq. (90) the trace over the driving field is evaluated as in Eq. (91), and we have

$$\wp_m^e(t_1,t_2,\ldots,t_m;[t,t+T])$$
$$=\mathrm{tr}\{\exp[(\mathcal{L}-\mathcal{S})(t+T-t_m)]\mathcal{S}\cdots\mathcal{S}\exp[(\mathcal{L}-\mathcal{S})(t_2-t_1)]\mathcal{S}\exp[(\mathcal{L}-\mathcal{S})(t_1-t)]\hat{\rho}(t-r_{AD}/c)\}\ ; \tag{104}$$

we have used $\mathrm{tr}_f(\mathcal{P}\hat{\rho}')=\mathrm{tr}_f(\hat{\rho}')=\hat{\rho}$.

Equation (104) defines the exclusive probability densities for photon emissions; they are the same as the exclusive probability densities for photoelectron counting sequences for unit detection efficiency [Eq. (75)]. To take the final step giving the $\wp_m(t_1,t_2,\ldots,t_m;[t,t+T])$ for $\eta'\neq1$, we must simply account for the fact that an emitted photon is detected with probability η', and is not detected with probability $(1-\eta')$. Let us look at the simplest example, the derivation of $\wp_1(t_1;[t,t+T])$. The one photoelectric count recorded at t_1 corresponds to the detection of a photon emitted at t_1-r_{AD}/c. This photon emission may be preceded during the interval $[t-r_{AD}/c,t_1-r_{AD}/c)$, and succeeded during the interval $(t_1-r_{AD}/c,t+T-r_{AD}/c]$, by any number of undetected emissions. Therefore,

$$\wp_1(t_1;[t,t+T])=\sum_{p=0}^{\infty}\sum_{q=0}^{\infty}\eta'(1-\eta')^{p+q}\int_0^{t+T-t_1}d\tau'_q\int_0^{\tau'_q}d\tau'_{q-1}\cdots\int_0^{\tau'_2}d\tau'_1$$
$$\times\int_0^{t_1-t}d\tau_p\int_0^{\tau_p}d\tau_{p-1}\cdots\int_0^{\tau_2}d\tau_1\wp_{p+q+1}^e(t+\tau_1,\ldots,t+\tau_p,t_1,t_1+\tau'_1,\ldots,t_1+\tau'_q;[t,t+T])\ . \tag{105}$$

After substituting for

$$\wp_{p+q+1}^e(t+\tau_1,\ldots,t+\tau_p,t_1,t_1+\tau'_1,\ldots,t_1+\tau'_q;[t,t+T])$$

from Eq. (104) and evaluating the sums using Eq. (55), we find

$$\wp_1(t_1;[t,t+T]) = \eta' \mathrm{tr}\{\exp[(\mathcal{L}-\mathcal{S})+(1-\eta')\mathcal{S}](t+T-t_1)\mathcal{S}\exp\{[(\mathcal{L}-\mathcal{S})+(1-\eta')\mathcal{S}](t_1-t)\}\hat{\rho}(t-r_{AD}/c)\}$$
$$= \eta' \mathrm{tr}\{\exp[(\mathcal{L}-\eta'\mathcal{S})(t+T-t_1)]\mathcal{S}\exp[(\mathcal{L}-\eta'\mathcal{S})(t_1-t)]\hat{\rho}(t-r_{AD}/c)\} \ . \quad (106)$$

Thus, $\wp_1^e(t_1;[t,t+T])$ is transformed into $\wp_1(t_1;[t,t+T])$ by the substitution $\mathcal{S}\to\eta'\mathcal{S}$. A more general calculation shows that Eq. (104) is transformed in the same manner into Eq. (75).

We have developed a picture of photon emission sequences that parallels the picture of photoelectron counting sequences in Secs. II and III. We are permitted the interpretation that photons are emitted, accompanied by the consequent reduction of the atomic state, without reference to the process of photoelectric detection. Detection is simply a matter of casting a net to catch emitted photons; they are caught randomly with probability η'. We may replace the statement, the atom returns to its ground state at t_0-r_{AD}/c due to photoelectric detection at t_0, by the statement, a photoelectric count is recorded at t_0 because the atom returned to its ground state (with the emission of a photon) at t_0-r_{AD}/c.

The exclusive probability densities for photon emission [Eq. (104)] can be written in the factorized form of Eq. (76). In particular, the distribution of waiting times between photon emissions is given by

$$w_e(\tau) \equiv w(\tau)_{\eta'=1}$$
$$= \langle + |\hat{\bar{\rho}}(\tau)| + \rangle \mathrm{tr}(e^{(\mathcal{L}-\mathcal{S})\tau}|-\rangle\langle -|) \ , \quad (107)$$

where

$$\hat{\bar{\rho}}(\tau) = \frac{e^{(\mathcal{L}-\mathcal{S})\tau}|-\rangle\langle -|}{\mathrm{tr}(e^{(\mathcal{L}-\mathcal{S})\tau}|-\rangle\langle -|)} \ ; \quad (108)$$

$\mathrm{tr}(e^{(\mathcal{L}-\mathcal{S})\tau}|-\rangle\langle -|)$ is the probability that no photon is emitted during the interval t_0-r_{AD}/c to $t+\tau-r_{AD}/c$ given a photon was emitted at t_0-r_{AD}/c, and $\hat{\bar{\rho}}(\tau)$ describes the reduced state of the atom at time $t_0+\tau-r_{AD}/c$ given a photon is emitted at t_0-r_{AD}/c and no photons are emitted during the interval t_0-r_{AD}/c to $t_0+\tau-r_{AD}/c$—the reduced state of the atom between photon emissions. According to our interpretation this state is reached dynamically, without any influence from the observer; although, of course, observation is necessary to know when each return to the ground state takes place. On the other hand, $\hat{\bar{\rho}}_{\eta'}(\tau)$ defined by Eq. (63) describes an atomic state inferred from imperfect information ($\eta'\neq 1$) about the photon emission sequences. It describes the observer's state of knowledge about the atom, given the available information is collected with less than perfect efficiency.

We are suggesting that photon emission sequences may be inserted as realized events lying behind the observed photoelectron counting sequences. We can do this because the source is an open system that loses energy *irreversibly* to the vacuum. The irreversibility effectively performs a continuous quantum measurement, without the need for a conscious observer to record the emitted photons. However, we must be careful not to go too far with our *realized* photon emissions; the observer does have a role to play. Photoelectron counting sequences are described by a classical stochastic process. Photon emission is a mixture of classical stochastics (describing emission times) and quantum mechanics. Source dynamics are certainly not described in their entirety by a classical stochastic process. We cannot even say that they decompose, unambiguously, into photon emission events that leave the atom in its ground state, connected by quantum evolution without emission. This is one permissible interpretation, one that is matched (by the choice of $\mathcal{S}$) to the behavior observed with a broadband detector. However, if the fluorescence is observed in a different manner, different behavior will be seen, behavior that does not fit the same interpretation of the emission events. Specifically, a narrowband detector designed to record photons within the frequency range of one Rabi sideband observes the atomic state reduced to one of the "dressed" states, a superposition of the ground and excited states. A different decomposition to that given by Eq. (97) can isolate the emission of these photons. In fact, it is possible to decompose $\mathcal{S}$ into four pieces, each describing one of the four types of transitions down the dressed state cascade,[29] each reducing the atomic state in its own way. The master equation [Eq. (57)] combines all available pictures of the photon emission process in a single *quantized* dissipative dynamical equation. As we expect with a quantum system, different measurement schemes see a different facet of a multifaceted quantum dynamic.

The observer's role is to select the facet of the source dynamics revealed by the method of observation. We should not confuse this role with the cause of the atomic state reduction. All potential reductions are incorporated within the source dynamics, and performed by the irreversible interaction with the vacuum, in the absence of an observer. The distinction is not purely semantic. This is made clear when we realize that there are other situations in which photoelectric detection does cause a collapse of the wave packet. This occurs, for example, when the detector monitors an otherwise closed system, extracting energy as it counts photons from a system that would not show a dissipative dynamic if the detector were turned off. Various authors have described photon counting of this type.[30–34] Most recently Srinivas and Davies discuss this approach as an example of the theory of continuous quantum measurement.[34] In fact the mathematical form of the decomposition of source dynamics outlined in this section parallels the language used by Srinivas and Davies to treat the interaction between a photoelectric detector and the field it measures. But it is irreversibility that provides the common ground, not

detector-field interaction. In the case of single-atom resonance fluorescence the irreversibility certainly does not originate in the presence of the observer's detector (one may, of course, take the viewpoint of absorber theory where it originates in the extended detector provided by the entire absorbing environment). Mandel has emphasized the distinction between open and closed systems in the relationship between the work of Srinivas and Davies and the Kelly-Kleiner theory of photoelectric detection.[35] He argued that the Kelly-Kleiner theory is appropriate for treating open systems. Using single-atom resonance fluorescence as an example, we have shown explicitly how Kelly-Kleiner theory accounts for the photoelectron counting statistics of a photoemissive source; with a modified interpretation it actually incorporates the language of Srinivas and Davies.

VI. SUMMARY AND CONCLUSIONS

We have studied photoelectron counting sequences and photon emission sequences for single-atom resonance fluorescence, emphasizing exclusive probability densities rather than the nonexclusive probability densities more commonly discussed. Photoelectron counting sequences are Markovian and stationary (after the first count) and are essentially characterized by the distribution of waiting times between photoelectric counts. We have calculated this waiting-time distribution, $w(\tau)$, allowing for arbitrary detection efficiency, both from the Kelly-Kleiner theory of photoelectric detection and from a modified set of Bloch equations for the atomic source.

For times much shorter than the average time between photoelectric counts, $w(\tau)$ is proportional to the degree of second-order temporal coherence $g_{ss}^{(2)}(\tau)$. Its full time dependence gives a clearer view of photon antibunching and sub-Poissonian counting statistics in resonance fluorescence than does $g_{ss}^{(2)}(\tau)$. In the limit of weak driving fields or low detection efficiency $w(\tau)$ approaches the exponential form for coherent light at all times except those that are much shorter than the mean time between photoelectric counts. The most dramatic nonclassical effects are seen for unit detection efficiency and moderate driving-field strengths, where $w(\tau)$ is peaked about the mean time between counts. Then photoelectric counts are recorded approximately regularly spaced in time.

We have formally related photoelectron counting sequences and photon emission sequences by calculating exclusive probability densities from two approaches—first from the theory of photoelectric detection, and then directly from the master equation describing the dissipative dynamics of the atomic source. We have argued that photon emission may be viewed as a realized process underlying observed photoelectron counting sequences. The detector simply records an emitted photon with probability η', or fails to record it with probability $(1-\eta')$. From this point of view the atomic state reduction associated with each photoelectric count is caused by the irreversible interaction of the atom with the multimode vacuum, not by the detection process itself. We have calculated the reduced state of the atom between photoelectric counts inferred by an observer monitoring the fluorescence with a detection efficiency η'.

Our discussion has been directed specifically at the problem of single-atom resonance fluorescence. However, the formal approach of Secs. III and V may be applied to analyze photoelectron counting sequences for other photoemissive sources. An important example is a radiating optical cavity with intracavity interaction. This general scheme includes such systems as the laser, optical bistability, and the parametric amplifier. Photoelectron counting sequences for a radiating cavity mode are not generally Markovian and the waiting-time distribution does not have such central importance. Future work will show whether exclusive probability densities provide a useful and practical tool for analyzing the emission and counting statistics for such a source.

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APPENDIX: AN ALGORITHM FOR COMPUTING $p(n,t,t+T)$

Inversion of the Laplace transform $\tilde{p}(n,s,t)$ requires the calculation of coefficients $C_{n,m}^{i}$, $i=1,2,3$, $m=0,1,\ldots,n$, in a partial fraction expansion of the right-hand side of Eq. (84). Then

$$p(n,t,t+T)=\eta'^{\,n}\sum_{m=0}^{n}\frac{(2\beta T)^m}{m!}\sum_{i=1}^{3}C_{n,m}^{i}\exp(\lambda_i,2\beta T)\ , \tag{A1}$$

where the $\lambda_i=s_i/2\beta$ are eigenvalues of the matrix

$$\underline{N}=\begin{bmatrix}-1 & 0 & \Omega/2\beta\\ 1-\eta' & 0 & -\Omega/2\beta\\ -\Omega/4\beta & \Omega/4\beta & -\frac{1}{2}\end{bmatrix}. \tag{A2}$$

$\underline{N}$ governs the time evolution of $p_0(\tau)$, $w(\tau)$, and $p_1(\tau|t)$. It is obtained by deleting the third row and third column from the matrix $\underline{M}$ defined by Eq. (72). We have

$$\tilde{p}_0(s)=\left[(\underline{N}-s\underline{I})^{-1}\begin{bmatrix}0\\1\\0\end{bmatrix}\right]_{11}+\left[(\underline{N}-s\underline{I})^{-1}\begin{bmatrix}0\\1\\0\end{bmatrix}\right]_{22}, \tag{A3}$$

$$\tilde{w}(s)=\eta'(2\beta)\left[(\underline{N}-s\underline{I})^{-1}\begin{bmatrix}0\\1\\0\end{bmatrix}\right]_{11}, \tag{A4}$$

$$\tilde{p}_1(s|t)=\eta'(2\beta)\left[(\underline{N}-s\underline{I})^{-1}\begin{bmatrix}\langle+|\hat{\rho}_t|+\rangle\\ \langle-|\hat{\rho}_t|-\rangle\\ \mathrm{Im}\langle+|\hat{\rho}_t|-\rangle\end{bmatrix}\right]_{11}, \tag{A5}$$

where $\hat{\rho}_t\equiv\hat{\rho}(t-r_{AD}/c)$ and $\underline{I}$ is the 3×3 identity matrix.

The coefficients $C_{n,m}^{i}$ appearing in Eq. (A1) can be computed from the eigenvalues and right and left eigenvectors of $\underline{N}$. We define matrices

$$\underline{R} \equiv (R_{ij}) = (\mathbf{r}_1 \mathbf{r}_2 \mathbf{r}_3) , \tag{A6}$$

and

$$\underline{L} \equiv (L_{ij}) = (\boldsymbol{l}_1 \boldsymbol{l}_2 \boldsymbol{l}_3)^T , \tag{A7}$$

with

$$\underline{L}\,\underline{R} = \underline{I} , \tag{A8}$$

where

$$\underline{N}\mathbf{r}_i = \lambda_i \mathbf{r}_i , \tag{A9}$$

and

$$\boldsymbol{l}_i^T \underline{N} = \lambda_i \boldsymbol{l}_i^T . \tag{A10}$$

The transformation $\underline{L}\,\underline{N}\,\underline{R} = \mathrm{diag}(\lambda_1, \lambda_2, \lambda_3)$ diagonalizes $\underline{N}$, and we may write

$$(\underline{N} - s\underline{I})^{-1} = \underline{R} \begin{bmatrix} (\lambda_1 - s)^{-1} & 0 & 0 \\ 0 & (\lambda_2 - s)^{-1} & 0 \\ 0 & 0 & (\lambda_3 - s)^{-1} \end{bmatrix} \underline{L} . \tag{A11}$$

The following algorithm for computing the $C^i_{n,m}$ is derived by substituting Eqs. (A3)–(A5) and (A11) into Eq. (84), and then collecting terms in $(s - \lambda_i)^{-(m+1)}$.

We define ($i = 1,2,3$)

$$\alpha_i = R_{1i} L_{i2} , \tag{A12}$$

$$\beta_i = (R_{1i} + R_{2i}) L_{i2} , \tag{A13}$$

$$\gamma_i = R_{1i}(L_{i1} \langle + | \hat{\rho}_t | + \rangle + L_{i2} \langle - | \hat{\rho}_t | - \rangle + L_{i3} \mathrm{Im} \langle + | \hat{\rho}_t | - \rangle) . \tag{A14}$$

Then

$$C^i_{0,0} = \beta_i \gamma_i / \alpha_i , \tag{A15}$$

$$C^i_{1,0} = \frac{\beta_i \gamma_j + \beta_j \gamma_i}{\lambda_i - \lambda_j} + \frac{\beta_i \gamma_k + \beta_k \gamma_i}{\lambda_i - \lambda_k} , \tag{A16}$$

$$C^i_{1,1} = \beta_i \gamma_i , \tag{A17}$$

and, for $n \geq 2$,

$$C^i_{n,0} = \sum_{r=0}^{n-1} \frac{\alpha_i C^j_{n-1,r} - (-1)^r \alpha_j C^i_{n-1,r}}{(\lambda_i - \lambda_j)^r} + \frac{\alpha_i C^k_{n-1,r} - (-1)^r \alpha_k C^i_{n-1,r}}{(\lambda_i - \lambda_k)^r} , \tag{A18}$$

$$C^i_{n,m} = \alpha_i C^i_{n-1,m-1} - \sum_{r=m}^{n-1} C^i_{n-1,r} \left[\frac{\alpha_j}{(\lambda_j - \lambda_i)^{r-m+1}} + \frac{\alpha_k}{(\lambda_k - \lambda_i)^{r-m+1}} \right] , \quad 1 \leq m \leq n-1 , \tag{A19}$$

$$C^i_{n,n} = \alpha_i C^i_{n-1,n-1} ; \tag{A20}$$

in Eqs. (A16), (A18), and (A19), (i,j,k) is a cyclic permutation of (1,2,3).

*Present address: Department of Physics, Miami University, Oxford, OH 45056.

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