Photoelectric Detection with Two-Photon Absorption*

A. K. JAISWAL AND G. S. AGARWAL
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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A formula for the photoelectron-counting distribution for a two-photon detector is derived quantum mechanically assuming that the ionising transitions in the atoms of the detector take place through the simultaneous absorption of two photons. It is assumed that the incident light is quasimonochromatic. It is shown that the distribution of the photoelectrons is given by the average of a Poisson distribution, the parameter of the distribution being proportional to the time integral of the square of the instantaneous light intensity. Counting distributions for the thermal (gaussian) light and for some models of laser light are obtained for the limiting case when the counting-time interval T is short compared to the coherence time T_c of the light. An approximate formula for arbitrary time intervals for the counting distribution of thermal light is also proposed.
INDEX HEADINGS: Coherence; Detection; Laser; Photoionization.

The problem of multiphoton absorption of radiation by atomic systems is one of the simplest processes involving nonlinear interaction of radiation with matter and has therefore been of considerable interest for a long time. The earliest work in this field is due to Goeppert-Mayer,[1] who considered the simultaneous absorption of two photons by an atomic system. But the interest in the field gathered momentum only with the availability of extremely intense light fluxes produced by lasers which made the experimental observation of these higher-order processes feasible. Since then, a large amount of experimental[2] and theoretical[3–5] work has been done in this field. The theory of N-photon photoionization was developed by Gold and Bebb[4] by the use of Nth-order time-dependent perturbation theory. Keldysh[5] has developed the theory by considering the phenomenon as the tunneling of a bound electron when it is subject to a static field. However, most of the works quoted above treat the problem mainly from the viewpoint of atomic physics without taking account of the statistical features of the radiation field. Only recently, the effect of the coherence properties of the field has been taken into account in calculating the transition rates for photoabsorption in 2-photon[6] and in N-photon[7] processes. It is found in these investigations[7] that the transition probability for N-photon absorption depends upon the (n,n)th-order correlation function of the electromagnetic field. In particular, the transition rate for absorption of thermal light is found to be higher than that for the laser light of the same intensity. [Editor's note: The authors insist on using the word "intensity," without definition, in the sense in which "irradiance" is defined in the OSA nomenclature, J. Opt. Soc. Am. **57**, 854 (1967).]

In the present paper, we develop the theory of photodetection in which ionizing transitions take place through the simultaneous absorption of two photons, each of which has energy less than the first ionizing potential of the atoms of the detector, but their combined energy exceeds the threshold of ionization. It is now well known that in the one-photon detector, in which the ionizing transitions take place through the absorption of one photon only, the statistical properties of the photoelectrons ejected from the surface of the photodetector provide information about the fluctuations[8–14] in the incident field. The basic result of the theory[12,13] as developed by Mandel is that the probability of registering n counts, in the counting-time interval T, is given by the Poisson transform of the probability density $P(W)$ of W

$$p(n;t,T)=\int \frac{W^n e^{-W}}{n!}P(W)dW, \tag{1}$$

where W is the time-integrated light intensity and is

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[1] M. Goeppert-Mayer, Ann. Physik **9**, 273 (1931).

[2] R. Braunstein and N. Ockman, Phys. Rev. **134**, A499 (1964); S. Yatsiv, W. G. Wagner, G. S. Picus, and F. J. McClung, Phys. Rev. Letters **15**, 614 (1965); E. M. Logothetics and P. L. Hartman, **18**, 581 (1967); and some of the references quoted in these papers.

[3] S. Kielich, Acta Phys. Polon. **30**, 393 (1966); R. Wallace, Mol. Phys. **11**, 457 (1966); and some of the references quoted therein.

[4] A. Gold and H. Barry Bebb, Phys. Rev. Letters **14**, 60 (1965); H. Barry Bebb and A. Gold, Phys. Rev. **143**, 1 (1966).

[5] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1966).

[6] B. R. Mollow, Phys. Rev. **175**, 1555 (1968) and some of the references quoted therein.

[7] G. S. Agarwal, Phys. Rev. **177**, 400 (1969).

[8] L. Mandel and E. Wolf, Rev. Mod. Phys. **37**, 231 (1965).

[9] L. Mandel in *Progress in Optics II*, E. Wolf, Ed. (North-Holland Publ. Co., Amsterdam, 1963), p. 181.

[10] J. A. Armstrong and A. W. Smith, in *Progress in Optics VI*, E. Wolf, Ed. (North-Holland Publ. Co., Amsterdam, 1967), p. 211, and some of the references quoted in this review article.

[11] J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics* (W. A. Benjamin, Inc., New York, 1968).

[12] L. Mandel, Proc. Phys. Soc. (London) **72**, 1037 (1958); **74**, 233 (1959).

[13] The original derivation of Mandel was based on semiclassical arguments. The corresponding quantum-mechanical derivation of the formula was given by R. J. Glauber, in *Quantum Optics and Electronics*, Les Houches 1964, C. Dewitt, A. Blandin, and C. Cohen-Tannoudji, Eds. (Gordon and Breach, New York, 1965), p. 65. See also P. L. Kelley and W. H. Kleiner, Phys. Rev. **136**, A316 (1964).

[14] E. Wolf and C. L. Mehta, Phys. Rev. Letters **13**, 705 (1964); G. Bédard, J. Opt. Soc. Am. **57**, 1201 (1967).

given by

$$W=\alpha\int_t^{t+T} I(t')dt', \tag{2}$$

α being the quantum efficiency parameter of the detector. The formula for $p(n;t,T)$ can be inverted[14] to give $P(W)$ in terms of $p(n;t,T)$. Thus information about the statistical properties of the field can be obtained from that of the photoelectrons.

In Sec. I, we use the second-order time-dependent perturbation theory to obtain the transition rate for two-photon ionization. Under reasonable assumptions about the atomic system and the bandwidth of the radiation field, we find that the counting distribution $p(n;t,T)$ is again of a similar form as in Eq. (1) except that the parameter W is now defined as the time integral of the square of the instantaneous light intensity. Because of the higher-order interaction involved, two-photon ionization would be expected to provide a more sensitive probe of the fluctuation of the field. As shown in Sec. II, it indeed turns out that for the short time intervals T, the kth factorial moment of the photo-counting distribution is related to the $2k$th moment of the field-intensity distribution. We also consider applications to the thermal light and to some models of laser light in this section. In the last section, an approximate formula for the counting distribution for thermal light, valid for arbitrary time interval T, is proposed.

I. PROBABILITY DISTRIBUTION OF PHOTOELECTRONS IN A TWO-PHOTON DETECTOR

In this section, we will first obtain the transition probability per unit time when a single atom of the detector interacts with the electromagnetic field incident upon it. The interaction hamiltonian between the radiation field and the atom is given by

$$\hat{H}_I(t)=-\frac{e}{mc}\hat{\mathbf{p}}(t)\cdot\hat{\mathbf{A}}(\mathbf{x},t)+\frac{e^2}{2mc^2}\hat{\mathbf{A}}^2(\mathbf{x},t), \tag{3}$$

where $\hat{\mathbf{A}}(\mathbf{x},t)$ is the vector-potential operator for the electromagnetic field and $\hat{\mathbf{p}}(t)$ is the momentum operator of the valence electron of the atom. In what follows, we shall work in the interaction picture and will use the dipole approximation, i.e., we will neglect the effect of the spatial variation of $\hat{\mathbf{A}}(\mathbf{x},t)$ over the dimensions of the atom. We will also neglect the effect of the term quadratic[15,16] in the vector potential of the electromagnetic field. We write the vector-potential operator as sum of positive and negative frequency parts, as usual,

$$\hat{\mathbf{A}}(\mathbf{x},t)\simeq\hat{\mathbf{A}}(t)=\hat{\mathbf{A}}^{(+)}(t)+\hat{\mathbf{A}}^{(-)}(t), \tag{4}$$

where

$$\mathbf{A}^{(+)}(t)=\sum_{k,s}\left(\frac{\hbar C}{kL^3}\right)^{\frac{1}{2}}\boldsymbol{\varepsilon}(k,s)\hat{a}_{k,s}e^{-i\omega_k t}. \tag{5}$$

In Eq. (5), $\hat{a}_{k,s}$ is the annihilation operator and $\boldsymbol{\varepsilon}(k,s)$ is the unit polarization vector associated with the mode (k,s) of the radiation field.

Let us assume that the atom is initially in the ground state $|0\rangle_a$ and let $|f\rangle_a$ be the final state of the atom after the absorption has taken place. Similarly, we denote by $|i\rangle_f$ and $|f\rangle_f$ the initial and the final states of the field. Then the probability that the system has made a transition to the final state $|f\rangle_a$ of the atom is given by

$$P(t)=\sum_{\{f\}f}|{}_f\langle f|{}_a\langle f|\hat{U}_I(t,t_0)|0\rangle_a|i\rangle_f|^2. \tag{6}$$

Here $\hat{U}_I(t,t_0)$ is the unitary time-development operator in the interaction picture and is given by the relation

$$\hat{U}_I(t,t_0)=\hat{T}\exp\left\{-\frac{i}{\hbar}\int_{t_0}^{t}\hat{H}_I(t')dt'\right\}, \tag{7}$$

where $\hat{T}$ is the Dyson time-ordering operator.

The probability of the two-photon absorption is obtained by the usual procedure, namely, by expanding the unitary operator $\hat{U}_I(t,t_0)$ into a series and retaining only that term in the expansion which contains a product of two-photon annihilation operators. This gives the following result for the transition probability to $|f\rangle_a$ by two-photon absorption in the time interval t_0 to $t_0+\Delta t$ during which the interaction is turned on

$$P^{(2)}(t)=\sum_{\{f\}f}\Big|\int_{t_0}^{t_0+\Delta t}dt_1\int_{t_0}^{t_1}dt_2\sum_{\mu_1\mu_2}\sum_{j}M_j(\mu_1,\mu_2)\times\exp\{-i(\omega_j-\omega_f)t_1+i\omega_j t_2\}\times{}_f\langle f|\hat{A}_{\mu_1}^{(+)}(t_1)\hat{A}_{\mu_2}^{(+)}(t_2)|i\rangle_f\Big|^2. \tag{8}$$

Here, we have separated the matrix element of the atomic operators from that of the field operators.

[15] N. V. Cohan and H. V. Hameka, Phys. Rev. Letters **16**, 478 (1966); Phys. Rev. **151**, 1076 (1966); See also R. Wallace, Phys. Rev. Letters **17**, 397 (1966); Mol. Phys. **11**, 457 (1966).

[16] The contribution of the A^2 term is also easily taken into account. It may be shown that the final result is still given by Eq. (30) except that the quantum-efficiency parameter α changes slightly. The new value of α is

$$\alpha=2\pi N\sum_{\{\mu\}}\sum_{jk}\frac{M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\epsilon_{\mu_1}\epsilon_{\mu_2}\epsilon_{\mu_3}^*\epsilon_{\mu_4}^*}{(\omega_j-\omega_0)(\omega_k-\omega_0)}\rho(2\omega_0)+2\pi N\sum_{\{\mu\}}\left(\frac{e^2}{2\hbar mc^2}\right)^2|\langle f|0\rangle|^2\epsilon_{\mu_1}^*\epsilon_{\mu_1}^*\epsilon_{\mu_2}\epsilon_{\mu_2}\rho(2\omega_0).$$

Here $\langle f|0\rangle$ represents the scalar product between the ground-state wavefunction and the final-state (free-electron) wavefunction.

We have also used the abbreviation

$$M_j(\mu_1,\mu_2)\equiv\left(\frac{e}{\hbar mc}\right)^2 {}_a\langle f|\hat{p}_{\mu_1}|j\rangle_{aa}\langle j|\hat{p}_{\mu_2}|0\rangle_a, \quad (9)$$

where $|0\rangle_a$, $|j\rangle_a$ and $|f\rangle_a$ stand for the initial, intermediate, and final state of the atomic system, respectively. The summation over the polarization indices has been denoted by $\sum_{\mu_1,\mu_2}$ in Eq. (8).

We must yet sum over the final states of the atom. Assuming that these states are in continuum and that $\rho(\omega_f)$ is the density of these states, we find that the transition probability per unit time is given by the relation

$$\Pi(t)=\frac{1}{\Delta t}\int_0^\infty P^{(2)}(t)\rho(\omega_f)d\omega_f. \quad (10)$$

Let us now assume that the density operator corresponding to the initial state of the field admits a diagonal-coherent-state representation[17]

$$\hat{\rho}_{if}=\int\Phi(\{\mathcal{V}_{k,s}\})|\{\mathcal{V}_{k,s}\}\rangle\langle\{\mathcal{V}_{k,s}\}|d^2(\{\mathcal{V}_{k,s}\}). \quad (11)$$

Here the coherent states $|\{\mathcal{V}_{k,s}\}\rangle$ are the eigenstates of the positive-frequency part of the field operator $\hat{\mathbf{A}}(\mathbf{x},t)$

$$\mathbf{A}^{(+)}(\mathbf{x},t)|\{\mathcal{V}_{k,s}\}\rangle=\mathbf{V}(\mathbf{x},t)|\{\mathcal{V}_{k,s}\}\rangle, \quad (12)$$

where

$$\mathbf{V}(\mathbf{x},t)\equiv\sum_{k,s}\left(\frac{\hbar c}{kL^3}\right)^{\frac{1}{2}}\boldsymbol{\varepsilon}(k,s)\mathcal{V}_{k,s}e^{-i\omega_k t}. \quad (13)$$

The eigenvalues $\mathbf{V}(\mathbf{x},t)$ are the so-called complex analytic signals.[18] They contain only the positive frequencies and can, therefore, be expanded,

$$\mathbf{V}(\mathbf{x},t)=\int_0^\infty A(\mathbf{x},\omega)e^{-i\omega t}d\omega. \quad (14)$$

Let us now assume that the field is initially in a coherent state. The more realistic situation is then taken into account by averaging over the generalized phase-space ensemble $\phi(\{\mathcal{V}_{k,s}\})$ at the end. We have chosen to work in the diagonal-coherent-state representation of the density operator of the field, because it brings out the close analogy between the quantum-mechanical and semiclassical treatment of the problem.[8,17] Many of the calculations that follow in this section are similar to the ones employed by Mandel, Sudarshan, and Wolf[19] in their semiclassical treatment of one-photon detector.

Summing over the final states of the field in Eq. (8) and using the relation (12), we obtain

$$P^{(2)}(t)=\sum_{\{\mu\}}\sum_{j,k}M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\int_{t_0}^{t_0+\Delta t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_0+\Delta t}dt_1'\int_{t_0}^{t_1'}dt_2'\,\exp\{-i(\omega_j-\omega_f)t_1+i\omega_j t_2+i(\omega_k-\omega_f)t_1' -i\omega_k t_2'\}V_{\mu_1}(t_1)V_{\mu_2}(t_2)V_{\mu_3}^*(t_1')V_{\mu_4}^*(t_2'). \quad (15)$$

When we insert the expansion (14) into Eq. (15), we get

$$P^{(2)}(t)=\sum_{\{\mu\}}\sum_{j,k}M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\int_0^\infty d\omega_1\int_0^\infty d\omega_2\int_0^\infty d\omega_1'\int_0^\infty d\omega_2' A_{\mu_1}(\omega_1)A_{\mu_2}(\omega_2)A_{\mu_3}^*(\omega_1')A_{\mu_4}^*(\omega_2')$$
$$\times\int_{t_0}^{t_0+\Delta t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_0+\Delta t}dt_1'\int_{t_0}^{t_1'}dt_2'\exp\{-it_1(\omega_j-\omega_f+\omega_1)+it_2(\omega_j-\omega_2)+i(\omega_k-\omega_f+\omega_1')t_1'-it_2'(\omega_k-\omega_2')\}. \quad (16)$$

We now perform the time integrals in Eq. (16). We first note that a finite lower limit of the integrals over t_2 and t_2' introduces nonresonant, rapidly oscillating terms which do not contribute towards the transition probability. Hence, we may replace these lower limits by $-\infty$ without affecting the results. Lower limits of the integrals over t_1 and t_1' are both taken to be $t=t_0$. This ensures that the final state was not occupied before the perturbation. On performing these time integrals, we obtain

$$P^{(2)}(t)=\sum_{\{\mu\}}\sum_{j,k}M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\int_0^\infty d\omega_1\int_0^\infty d\omega_2\int_0^\infty d\omega_1'\int_0^\infty d\omega_2' A_{\mu_1}(\omega_1)A_{\mu_2}(\omega_2)A_{\mu_3}^*(\omega_1')A_{\mu_4}^*(\omega_2')(t)^2$$
$$\times(\Delta t)^2\left\{\sin[\tfrac{1}{2}(\omega_1+\omega_2-\omega_f)\Delta t]\Big/[\tfrac{1}{2}(\omega_1+\omega_2-\omega_f)\Delta t]\sin[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_f)\Delta t]\Big/[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_f)\Delta t]\right\}$$
$$\times\exp\left\{i(\omega_1'+\omega_2'-\omega_1-\omega_2)\left(t_0+\frac{\Delta t}{2}\right)\right\}\Big/\left\{(\omega_j-\omega_2)(\omega_k-\omega_2')\right\}. \quad (17)$$

[17] E. C. G. Sudarshan, Phys. Rev. Letters **10**, 277 (1963). A detailed discussion of the diagonal representation is given in Ref. 11. See also R. J. Glauber, Phys. Rev. **131**, 2766 (1963).
[18] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Ltd., Oxford, England, 1965), 3rd ed., Ch. X.
[19] L. Mandel, E. C. G. Sudarshan, and E. Wolf, Proc. Phys. Soc. (London) **84**, 435 (1964).

To obtain $\Pi(t)$, we integrate $P^{(2)}(t)$, given by Eq. (17), over the density of final states of the atom. Let us first consider the integral over ω_f

$$I=\int_0^\infty d\omega_f\rho(\omega_f)\left\{\sin[\tfrac{1}{2}(\omega_1+\omega_2-\omega_f)\Delta t]\Big/[\tfrac{1}{2}(\omega_1+\omega_2-\omega_f)\Delta t]\sin[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_f)\Delta t]\Big/[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_f)\Delta t]\right\}. \quad (18)$$

We will now assume that the incident radiation is quasimonochromatic, i.e., its effective bandwidth Γ is small compared with the midfrequency ω_0. It is then possible to choose $1/\Delta t$ small compared to any frequency ω inside the narrow band of the radiation, but large compared with the difference $(\omega_1+\omega_2)-(\omega_1'+\omega_2')$, i.e.,

$$(\omega_1+\omega_2)-(\omega_1'+\omega_2')\ll\frac{1}{\Delta t}\ll 2\omega_0. \quad (19)$$

We then note that the expression inside the curly brackets in Eq. (18) does not contribute unless $\omega_1+\omega_2-\omega_f\lesssim\Gamma$ and $\omega_1'+\omega_2'-\omega_f\lesssim\Gamma$. For such a short range of integration over ω_f, the density of states $\rho(\omega_f)$ does not vary significantly and we may replace it by $\{\rho(\omega_1+\omega_2)\rho(\omega_1'+\omega_2')\}^{\frac{1}{2}}$. The integral (18), under the conditions dictated by inequality (19), reduces then to

$$I=\frac{2\pi}{\Delta t}\{\rho(\omega_1+\omega_2)\rho(\omega_1'+\omega_2')\}^{\frac{1}{2}} \times\sin[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_1-\omega_2)\Delta t]/[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_1-\omega_2)\Delta t]. \quad (20)$$

Using Eqs. (10), (17), (20), and the identity

$$\sin[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_1-\omega_2)\Delta t]/[\tfrac{1}{2}(\omega_1'+\omega_2'-\omega_1-\omega_2)\Delta t] =\frac{1}{\Delta t}\exp\left\{-i(\omega_1'+\omega_2'-\omega_1-\omega_2)\frac{\Delta t}{2}\right\} \times\int_0^{\Delta t}\exp\{i(\omega_1'+\omega_2'-\omega_1-\omega_2)\tau\}d\tau, \quad (21)$$

we obtain

$$\Pi(t)=\frac{2\pi}{\Delta t}\int_0^{\Delta t}d\tau\sum_{\{\mu\}}\sum_{j,k}M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\int_0^\infty d\omega_1\int_0^\infty d\omega_2\int_0^\infty d\omega_1'\int_0^\infty d\omega_2'\,A_{\mu_1}(\omega_1)A_{\mu_2}(\omega_2)A_{\mu_3}^*(\omega_1')A_{\mu_4}^*(\omega_2') \times\frac{\{\rho(\omega_1+\omega_2)\rho(\omega_1'+\omega_2')\}^{\frac{1}{2}}}{(\omega_j-\omega_2)(\omega_k-\omega_2')}\exp\{i(\omega_1'+\omega_2'-\omega_1-\omega_2)(t+\tau)\}. \quad (22)$$

Because of the assumption of quasimonochromaticity, $\{\rho(\omega_1+\omega_2)\rho(\omega_1'+\omega_2')\}^{\frac{1}{2}}$ does not vary significantly over the narrow band of frequencies and can be replaced by its value at the mid-frequency, i.e., $\rho(2\omega_0)$. Moreover, according to our assumption, none of the frequencies inside the narrow band of radiation is close to the intermediate frequencies. Hence, with similar arguments, the atomic response function in the denominator of Eq. (22) can be replaced by its value at the mid-frequency and we obtain

$$\Pi(t)=2\pi\left\{\sum_{\{\mu\}}\sum_{j,k}\frac{M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)\rho(2\omega_0)}{(\omega_j-\omega_0)(\omega_k-\omega_0)}\right\}\frac{1}{\Delta t}\int_0^{\Delta t}V_{\mu_1}(t+\tau)V_{\mu_2}(t+\tau)V_{\mu_3}^*(t+\tau)V_{\mu_4}^*(t+\tau)d\tau. \quad (23)$$

So far, we have considered only the interaction of a single atom with the incident radiation. Let us now consider the realistic situation where a plane-wave radiation field is incident on an extended detector which is in the form of a thin photoelectric layer. If we assume that the different atoms of the detector are independent and that the states are not appreciably depopulated, the transition probability is $N\times\Pi(t)$, where N is the number of atoms in the detector.

We will now write the vector complex field in the following way[20]

$$\mathbf{V}(t)=\boldsymbol{\varepsilon}\mathcal{V}(t), \quad (24)$$

where $\boldsymbol{\varepsilon}$ is a unit vector and $\mathcal{V}(t)$ is a complex scalar function. It can also be shown that under our assump-

[20] The representation of V in the form Eq. (24) assumes that the incident light is plane polarized. However, the results are easily generalized to the case of partially polarized light. If the incident light is of thermal origin, then as well known we may consider it as the superposition of two linearly polarized, statistically independent components with average intensities equal to $\frac{1}{2}(1+P)\langle I\rangle$ and $\frac{1}{2}(1-P)\langle I\rangle$, respectively, where P is the degree of polarization. It is also assumed that the condition of cross-spectral purity is satisfied. In this case, the final result (30) remains unchanged. (See also Sec. IIIA.) For the partially polarized light of nonthermal origin, the expression for the photoelectron counting distribution may be shown to be

$$p(n,T)=\int\Phi(\{\mathcal{V}_{ks}\})\frac{[W(\{\mathcal{V}_{ks}\})]^n\exp\{-W(\{\mathcal{V}_{ks}\})\}}{n!}d^2(\{\mathcal{V}_{ks}\}),$$

where

$$W(\{\mathcal{V}_{ks}\})=\sum_{\{\mu\}}\alpha_{\mu_1\mu_2\mu_3\mu_4}\int_0^T dt\,V_{\mu_1}(t)V_{\mu_2}(t)V_{\mu_3}^*(t)V_{\mu_4}^*(t),$$

and

$$\alpha_{\mu_1\mu_2\mu_3\mu_4}=2\pi N\sum_{jk}\frac{M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_4)}{(\omega_j-\omega_0)(\omega_k-\omega_0)}\rho(2\omega_0).$$

tion $\Delta t \ll 1/\Gamma$, we may write

$$\frac{1}{\Delta t}\int_0^{\Delta t}\{\mathcal{V}^*(t+\tau)\mathcal{V}(t+\tau)\}^2 d\tau \simeq \{\mathcal{V}^*(t)\mathcal{V}(t)\}. \quad (25)$$

Using Eqs. (24) and (25) in Eq. (23), we obtain for the probability of photoemission of one electron by the simultaneous absorption of two photons in the time interval t, $t+\Delta t$

$$P(t)\Delta t = \alpha\{\mathcal{V}^*(t)\mathcal{V}(t)\}^2\Delta t, \quad (26)$$

where

$$\alpha = 2N\pi\sum_{\{\mu\}}\sum_{j,k}\frac{M_j(\mu_1,\mu_2)M_k^*(\mu_3,\mu_2)\epsilon_{\mu_1}\epsilon_{\mu_2}\epsilon_{\mu_3}^*\epsilon_{\mu_4}^*\rho(2\omega_0)}{(\omega_j-\omega_0)(\omega_k-\omega_0)}$$

is a constant and may be called the quantum efficiency of two-photon detection.

Now we note that $\mathcal{V}^*(t)\mathcal{V}(t) = \mathbf{V}(t)\cdot\mathbf{V}(t)$ can be identified as the instantaneous intensity of the radiation field if we assume that all of the modes in the expansion (13) are similarly polarized [see Eq. (24)]. We then obtain

$$P(t)\Delta t = \alpha I^2(t)\Delta t. \quad (27)$$

If we consider the probability of photoemission as statistically independent for different time intervals, it readily follows[21] that $p(n;t,T)$ is given by

$$p(n;t,T) = \frac{W^n e^{-W}}{n!}, \quad (28)$$

where

$$W = \alpha\int_t^{t+T} I^2(t')dt'. \quad (29)$$

To obtain the probability distribution for photocounts, corresponding to the density operator of the field given by Eq. (11), we must average Eq. (28) over the phase-space distribution function of the field. We thus obtain[16,20]

$$p(n;t,T) = \int \phi(\{\mathcal{V}_{k,s}\}) \times \frac{[W(\{\mathcal{V}_{k,s}\})]^n e^{-W(\{\mathcal{V}_{k,s}\})}}{n!} d^2(\{\mathcal{V}_{k,s}\}), \quad (30)$$

which may be rewritten as

$$p(n;t,T) = \int_0^\infty \frac{W^n e^{-W}}{n!} P(W)dW, \quad (31)$$

where

$$P(W) = \int \phi(\{\mathcal{V}_{k,s}\})\delta\left(W - \alpha\int_t^{t+T} I^2(t')dt'\right) \times d^2(\{\mathcal{V}_{k,s}\}). \quad (32)$$

[21] See Ref. 9, Appendix B. Similar arguments are summarized in L. Mandel, Phys. Rev. 152, 438 (1966).

II. COUNTING DISTRIBUTIONS FOR $T \ll T_c$

In this section, we will discuss the photocounting distributions for a few cases of physical interest, namely for narrow-band gaussian light and for laser light. We will assume, in this section, that the counting-time interval T is much shorter than the coherence time T_c of the light falling on the detector. The counting-time intervals for which the above approximation holds can be obtained in practice for laser light and for pseudothermal light. Under this approximation, we may write

$$W = \alpha\int_t^{t+T} I^2(t')dt' \simeq \alpha T I^2(t). \quad (33)$$

Assuming that the radiation field is stationary, we then obtain, for the photocounting distribution,

$$p(n,T) = \int \frac{(\alpha I^2 T)^n}{n!} e^{-\alpha I^2 T} P(I)dI. \quad (34)$$

From Eq. (34), we may obtain the factorial moments $\langle n^{[k]}\rangle$. They are given by

$$\langle n^{[k]}\rangle = \sum_n n(n-1)\cdots(n-k+1)p(n;T) = (\alpha T)^k\langle I^{2k}\rangle. \quad (35)$$

Equation (35) implies that the kth factorial moment of the photocount distribution is proportional to the $2k$th intensity moment of the field.

We will now consider some typical cases.

A. Narrow-Band Gaussian Light

The intensity distribution for completely polarized gaussian light is given by

$$P(I) = \frac{1}{\langle I\rangle} e^{-I/\langle I\rangle}. \quad (36)$$

Substituting Eq. (36) into Eq. (34), we obtain

$$p(n,T) = \frac{2n!}{n!2^n}\frac{e^{\frac{1}{4}\langle n\rangle}}{\langle n\rangle^{\frac{1}{2}}} D_{-(2n+1)}(\langle n\rangle^{\frac{1}{2}}). \quad (37)$$

Here $\langle n\rangle = \alpha\langle I^2\rangle T$ is the average number of counts registered in the interval T, and $D_p(Z)$ are the parabolic-cylinder functions.[22] In obtaining relation (37), we have made use of the integral representation for the parabolic-cylinder functions[22]

$$D_p(Z) = \frac{1}{\Gamma(-p)}\int_0^\infty x^{-p-1}\exp\left\{-\frac{x^2}{2} - Zx - \frac{Z^2}{4}\right\}dx, \quad \mathrm{Re}\,p<0. \quad (38)$$

[22] See, for example, I. S. Gradshteyn and I. M. Ryzhik, *Table of Integrals, Series and Products* (Academic Press Inc., New York, 1965), p. 1064.

The result Eq. (37) should be compared with the Bose–Einstein distribution obtained for the case of the one-photon detector.

When the light is partially polarized, the intensity distribution is given by[23]

$$P(I)=\frac{1}{P\langle I\rangle}\left\{\exp\left\{\frac{-2I}{\langle I\rangle(1+P)}\right\}-\exp\left\{\frac{-2I}{\langle I\rangle(1-P)}\right\}\right\}, \quad (39)$$

where P is the degree of polarization. From Eqs. (34) and (39), we then obtain

$$p(n,T)=\frac{2n!}{n!2^n}\left(\frac{4\langle n\rangle P^2}{3+P^2}\right)^{\frac{1}{2}}\left\{D_{-(2n+1)}\left[\frac{1}{(1+P)}\left(\frac{3+P^2}{\langle n\rangle}\right)^{\frac{1}{2}}\right]\right.$$
$$\times\exp\left\{\frac{3+P^2}{4\langle n\rangle(1+P)^2}\right\}-D_{-(2n+1)}\left[\frac{1}{(1-P)}\left(\frac{3+P^2}{\langle n\rangle}\right)^{\frac{1}{2}}\right]$$
$$\left.\times\exp\left\{\frac{3+P^2}{4\langle n\rangle(1-P)^2}\right\}\right\}, \quad (40)$$

where

$$\langle n\rangle=\alpha T\langle I\rangle^2\left(\frac{3+P^2}{2}\right). \quad (41)$$

B. Amplitude Stabilized Laser

In this case, there are no intensity fluctuations and hence the probability density $P(I)$ is a delta function

$$P(I)=\delta(I-\langle I\rangle). \quad (42)$$

The probability distribution for the photoelectrons is therefore a Poisson distribution,

$$p(n,T)=\frac{\langle n\rangle^n e^{-\langle n\rangle}}{n!}, \quad (43)$$

where

$$\langle n\rangle=\alpha T\langle I\rangle^2. \quad (44)$$

C. Output of a Van der Pohl Oscillator

In the case of one-photon detector, it has been observed experimentally that the photocounting distribution for laser light is not strictly poissonian. It has been found[24,25] that this discrepancy may be removed if we assume that the intensity distribution for laser light is given by

$$P(I)=\frac{2}{\pi I_0}\left[\frac{e^{-\omega^2}}{1+\mathrm{err}(\omega)}\right]\exp\left\{\frac{-I^2}{\pi I_0^2}+\frac{2\omega I}{\sqrt{\pi}I_0}\right\}, \quad I\geqslant 0. \quad (45)$$

This expression was obtained by Risken[26] assuming a Van der Pohl oscillator model for a laser. Here I_0 is the average light intensity at threshold and the parameter ω varies from large negative values to large positive values as the laser is brought from well below threshold to well above threshold. Straightforward calculations give for the counting distribution for a two-photon detector, when the intensity distribution is given by Eq. (45),

$$p(n,T)=\frac{2n!}{n!2^n}\sqrt{\frac{2}{\pi}}\frac{\sigma_0^n}{(1+\sigma_0)^{n+\frac{1}{2}}}$$
$$\times\exp\left\{-\frac{\omega^2}{2}\left(\frac{1+2\sigma_0}{1+\sigma_0}\right)\right\}\Big/[1+\mathrm{err}(\omega)].$$
$$\times D_{-(2n+1)}\left[-\omega\Big/\left(\frac{1+\sigma_0}{2}\right)^{\frac{1}{2}}\right], \quad (46)$$

where

$$\sigma_0=\pi\langle n_0\rangle=\pi\alpha T I_0^2. \quad (47)$$

The factorial moments, in this case, are given by

$$\langle n^{[k]}\rangle=\frac{2k!\sigma_0^k}{2^k}\sqrt{\frac{2}{\pi}}\left\{\exp\left(-\frac{\omega^2}{2}\right)\Big/1+\mathrm{err}(\omega)\right\}.$$
$$\times D_{-(2k+1)}(-\omega\sqrt{2}). \quad (48)$$

III. APPROXIMATE COUNTING DISTRIBUTIONS FOR ARBITRARY TIME INTERVALS

It is very difficult to obtain an exact expression for the counting distribution for an arbitrary value of the counting-time interval T. In the case of thermal light, it is, however, possible to give an approximate formula, which is in good agreement with the exact expression for cases when T is either much shorter than, or much greater than, the coherence time T_c of the light beam. For the case of the one-photon detector, such an approximate formula was proposed by Mandel[12] and subsequently Bédard, Chang, and Mandel[27] showed that this formula is in fairly good agreement with the exact results. For a two-photon detector, following arguments similar to those of Mandel[12] (see also Ref. 28), we propose the following distribution for W

$$P(W)=\frac{a^{2k}}{2\Gamma(2k)}W^{k-1}e^{-aW^{\frac{1}{2}}}, \quad (49)$$

where the parameters a and k are to be chosen so that Eq. (49) gives the first two moments of W correctly. To determine a and k, we note from Eq. (29) that the

[23] See, for example, Ref. 8, Eq. (4.41).

[24] A. W. Smith and J. A. Armstrong, Phys. Rev. Letters **16**, 1169; R. F. Chang, R. W. Detenbeck, V. Korenman, C. O. Alley, Jr., and V. Hochuli, Phys. Letters **25A**, 272 (1967).

[25] G. Bedard, Phys. Letters **24A**, 613 (1967).

[26] H. Risken, Z. Physik **186**, 85 (1965).

[27] G. Bédard, J. C. Chang, and L. Mandel, Phys. Rev. **160**, 1496 (1967).

[28] S. O. Rice, Bell System Tech. J. 24, 46 (1945).

mean and variance of W are given by

$$\langle W\rangle=\alpha T\langle I^2\rangle, \tag{50}$$

and

$$\langle(\Delta W)^2\rangle=\alpha^2\int_0^T\int_0^T dt_1dt_2\{\langle I^2(t_1)I^2(t_2)\rangle-\langle I^2(t_1)\rangle\langle I^2(t_2)\rangle\}. \tag{51}$$

If the field incident upon the two-photon detector is gaussian, the moment theorem for gaussian random processes can be used to simplify Eqs. (50) and (51). It may be shown that

$$\langle I^2\rangle=2\langle I\rangle^2 \tag{52}$$

and

$$\langle I^2(t_1)I^2(t_2)\rangle-\langle I^2(t_1)\rangle\langle I^2(t_2)\rangle=\langle I\rangle^4\{16|\gamma(t_1-t_2)|^2+4|\gamma(t_1-t_2)|^4\}, \tag{53}$$

where $\gamma(t_1-t_2)$ is the normalized complex degree of coherence.[17] From Eqs. (51) and (53), it can be shown that

$$\langle(\Delta W)^2\rangle=4\alpha^2T^2\frac{\xi(T)}{T}\langle I\rangle^4, \tag{54}$$

where

$$\xi(T)=\frac{1}{2}\int_0^T\left(1-\frac{\tau}{T}\right)\{16|\gamma(\tau)|^2+4|\gamma(\tau)|^4\}d\tau. \tag{55}$$

From Eqs. (49)–(55), we find that a and k are given by

$$k=\frac{1}{4}\left[\left(\frac{4T}{\xi(T)}-1\right)+\left\{16\left(\frac{T}{\xi(T)}\right)^2+16\frac{T}{\xi(T)}+1\right\}^{\frac{1}{2}}\right] \tag{56}$$

and

$$a=\left\{\frac{2k(2k+1)}{\langle W\rangle}\right\}^{\frac{1}{2}}. \tag{57}$$

From Eqs. (56) and (57), it is easily seen that for $T\ll T_c$, the limiting values of a and k are

$$k=\tfrac{1}{2}\quad\text{and}\quad a=\left(\frac{2}{\langle W\rangle}\right)^{\frac{1}{2}}. \tag{58}$$

It therefore follows from Eq. (49) that

$$P(W)=(2W\langle W\rangle)^{-\frac{1}{2}}e^{-(2W/\langle W\rangle)^{\frac{1}{2}}}, \tag{59}$$

which is the correct distribution for $T\ll T_c$. In the same way, it can be shown that $P(W)$ defined by Eq. (49) tends to a delta function, $\delta(W-\langle W\rangle)$ when $T\gg T_c$. This is what we would expect, because for large integration time T, the fluctuations of W are smoothed out.

We may now obtain the approximate expression for the photocounting distribution by substituting Eq. (49) in Eq. (31),

$$p(n;T)=\frac{a^{2k}\Gamma(2n+2k)}{2^{n+k}n!\Gamma(2k)}e^{a^2/8}D_{-(2n+2k)}(a/\sqrt{2}). \tag{60}$$

The mean and variance of the photocounting distributions are given by

$$\langle n\rangle=\langle W\rangle$$

and

$$\langle(\Delta n)^2\rangle=\langle n\rangle\left\{1+\frac{\langle n\rangle\xi(T)}{T}\right\}, \tag{61}$$

which are obviously exact.

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