Off-diagonal density matrix for single-beam two-photon absorbed light

1978 J. Phys. A: Math. Gen. 11 435

(http://iopscience.iop.org/0305-4470/11/2/018)

J. Phys. A: Math. Gen., Vol. 11, No. 2, 1978. Printed in Great Britain. © 1978

ADDENDUM

Off-diagonal density matrix for single-beam two-photon absorbed light

H D Simaan† and R Loudon‡

† Physics Department, College of Science, Al-Mustansiriyah University, Baghdad, Iraq
‡ Physics Department, Essex University, Colchester CO4 3SQ, UK

Received 14 September 1977

Abstract. We derive general expressions for the time dependence of off-diagonal matrix elements of the density operator for single-mode light subjected to two-photon absorption. In conjunction with previous results for the diagonal matrix elements, they enable all measurable properties of the light to be calculated. The case of electric-field measurements is briefly considered.

1. Introduction

The present paper is an addendum to that of Simaan and Loudon (1975) which considered the time development of the statistical properties of a single beam of light subjected to two-photon absorption. The discussion was there limited to the diagonal elements of the density operator and it was shown that for various kinds of initial light beam, the light eventually becomes 'antibunched', that is, its degree of second order coherence falls below the value unity (see also Chandra and Prakash 1970). There has been recent interest in the properties of antibunched light (see Loudon 1976 for a review) and the main aim of the present extension is a fuller study of such light. We here derive the time dependence of the off-diagonal matrix elements of the density operator for two-photon absorbed light, thus providing a complete specification for all measurable properties of the light. The results are applied to one of the simplest off-diagonal properties, namely the electric field of the light beam.

2. Hamiltonian and master equation

The basic system considered and much of the notation used are the same as in Simaan and Loudon (1975) and need only be briefly specified. The Hamiltonian for the coupled light and the N non-interacting two-level atoms in two-photon resonance is (Shen 1967)

$$\hat{H} = \hbar\omega\hat{a}^\dagger\hat{a} + \tfrac{1}{2}\hbar\omega_0\sum_i(\hat{c}^\dagger_{2i}\hat{c}_{2i} - \hat{c}^\dagger_{1i}\hat{c}_{1i}) + \hbar\sum_i(K\hat{c}^\dagger_{2i}\hat{c}_{1i}\hat{a}\hat{a} + K^*\hat{a}^\dagger\hat{a}^\dagger\hat{c}^\dagger_{1i}\hat{c}_{2i}), \tag{1}$$

where ω and ω_0 $(=2\omega)$ are the frequencies of the light and the atomic transition, $\hat{a}^\dagger$ and $\hat{a}$ are photon creation and destruction operators, and the c operators refer to the ground state $|1\rangle$ and excited state $|2\rangle$ of the ith atom.

It is assumed here that almost all the N atoms are maintained in their ground states by some external influence and that two-photon emission can therefore be ignored. Standard techniques (Shen 1967, McNeil and Walls 1974) then lead to the following master equation for the reduced density operator $\hat{\rho}$ of the light field:

$$\mathrm{d}\hat{\rho}/\mathrm{d}t = N(J/2)(2\hat{a}\hat{a}\hat{\rho}\hat{a}^{\dagger}\hat{a}^{\dagger} - \hat{a}^{\dagger}\hat{a}^{\dagger}\hat{a}\hat{a}\hat{\rho} - \hat{\rho}\hat{a}^{\dagger}\hat{a}^{\dagger}\hat{a}\hat{a}), \tag{2}$$

where J is proportional to $|K|^2$. The off-diagonal matrix elements of the density operator in the Fock representation thus satisfy

$$\mathrm{d}\rho_{n,n'}/\mathrm{d}\tau = [(n+1)(n+2)(n'+1)(n'+2)]^{1/2}\rho_{n+2,n'+2} - \tfrac{1}{2}[n(n-1)+n'(n'-1)]\rho_{n,n'} \tag{3}$$

where

$$\tau = NJt. \tag{4}$$

The solution of these equations for the diagonal case $n = n'$ has been extensively treated by Simaan and Loudon (1975).

It is convenient in the off-diagonal case to set

$$\rho_{n,n'}(\tau) = \rho_{n,n+\mu}(\tau) = [n!/(n+\mu)!]^{1/2}\psi_n(\mu, \tau), \tag{5}$$

where μ is restricted to positive integers and denotes the degree of off-diagonality. The inverse matrix elements are given by

$$\rho_{n+\mu,n}(\tau) = \rho^{*}_{n,n+\mu}(\tau). \tag{6}$$

The transformed matrix elements ψ_n satisfy an equation obtained from (3)

$$\mathrm{d}\psi_n/\mathrm{d}\tau = (n+1)(n+2)\psi_{n+2} - [n(n-1) + \mu n + \tfrac{1}{2}\mu(\mu-1)]\psi_n, \tag{7}$$

which is now free of square-root factors. Note that the equation only connects matrix elements with equal degrees of off-diagonality.

We give particular attention to beams which are initially in a coherent state, where the matrix elements are (Loudon 1973)

$$\rho_{n,n'}(0) = \exp(-|\alpha|^2)\alpha^{n}\alpha^{*n'}/(n!n'!)^{1/2} \tag{8}$$

and the mean photon number is $|\alpha|^2$.

3. Steady-state solution

After a sufficiently long period of time has elapsed, the photon system settles down into a steady state in which all the rates of change (7) are zero. Summation of (7) over n then gives

$$\sum_n \mu(2n+\mu-1)\psi_n(\mu, \infty) = 0. \tag{9}$$

Noting from (7) that all the transformed matrix elements have the same sign in the steady state, we deduce from (9) that the only off-diagonal matrix element which can differ from zero is $\psi_0(1, \infty)$. The diagonal elements ($\mu = 0$), treated by Simaan and Loudon (1975), can be non-zero only for $n = 0$ and 1.

For the case $\mu = 1$, it is not difficult to prove from the equation of motion (7) that

$$(\mathrm{d}/\mathrm{d}\tau)\sum_n^{\text{even}} \{n!/2^n[(\tfrac{1}{2}n)!]^2\}\psi_n(1, \tau) = 0. \tag{10}$$

The existence of this constant of the motion provides an expression for the steady-state off-diagonal matrix element in terms of the initial matrix elements

$$\rho_{0,1}(\infty)=\psi_0(1,\infty)=\sum_n^{\text{even}}\{n!/2^n[(\tfrac{1}{2}n)!]^2\}\psi_n(1,0). \tag{11}$$

All other $\mu=1$ matrix elements decay from their initial values to become zero in the steady state.

Thus in terms of the original notation, the only matrix elements which can be non-zero in the steady state are $\rho_{1,1}$, $\rho_{0,0}$, $\rho_{0,1}$ and $\rho_{1,0}$. These conclusions are borne out by the results of the following section for the general time dependence of the matrix elements.

Consider as an example the initially coherent state described by (8). We put

$$\alpha=|\alpha|\exp \mathrm{i}\theta \tag{12}$$

and (5) gives

$$\psi_n(1,0)=(n+1)^{1/2}\rho_{n,n+1}(0)=\exp(-|\alpha|^2-\mathrm{i}\theta)|\alpha|^{2n+1}/n!. \tag{13}$$

The steady-state matrix element from (11) is

$$\rho_{0,1}(\infty)=\exp(-|\alpha|^2-\mathrm{i}\theta)|\alpha|I_0(|\alpha|^2), \tag{14}$$

where

$$I_0(|\alpha|^2)=\sum_{m=0}^{\infty}(\tfrac{1}{2}|\alpha|^2)^{2m}/(m!)^2 \tag{15}$$

is a modified Bessel function of the first kind. Its asymptotic form for $|\alpha|^2\gg 1$ is

$$I_0(|\alpha|^2)\to\exp(|\alpha|^2)/(2\pi)^{1/2}|\alpha|, \tag{16}$$

leading to

$$\rho_{0,1}(\infty)=\exp(-\mathrm{i}\theta)/(2\pi)^{1/2} \qquad (|\alpha|^2\gg 1) \tag{17}$$

for initially intense coherent light. The two steady-state diagonal matrix elements have the value 0·5 in the same limit (Simaan and Loudon 1975).

An initially chaotic light beam has zero off-diagonal matrix elements of the density operator at all times.

4. General-time solution

The equation of motion (7) is solved by the use of a generating function defined as

$$G(y,\mu,\tau)=\sum_{n=0}^{\infty}y^n\psi_n(\mu,\tau). \tag{18}$$

On multiplication of both sides of (7) by y^n and summation over n we obtain

$$\partial G/\partial\tau=(1-y^2)(\partial^2G/\partial y^2)-\mu y(\partial G/\partial y)-\mu(\mu-1)G. \tag{19}$$

If G is known, the transformed matrix elements can be found from

$$\psi_n(\mu,\tau)=(n!)^{-1}(\partial^nG/\partial y^n)_{y=0}. \tag{20}$$

The equation for the generating function is solved by the method of separation of the variables, as in the diagonal case (McNeil and Walls 1974, Simaan and Loudon 1975). The solution has the form

$$G(y, \mu, \tau) = \sum_{k=0}^{\infty} A_k^\sigma C_k^\sigma(y) \exp(-\lambda_k \tau), \tag{21}$$

where $C_k^\sigma(y)$ is a Gegenbauer polynomial with

$$\sigma = \tfrac{1}{2}(\mu - 1) \tag{22}$$

and

$$\lambda_k = k(k + \mu - 1) + \tfrac{1}{2}\mu(\mu - 1). \tag{23}$$

The coefficients A_k^σ are determined from the initial conditions, and using standard properties of the Gegenbauer polynomials, as in the diagonal case, we find

$$A_k^\sigma = \frac{(k+\sigma)\Gamma(\sigma)}{2^k \pi^{1/2}} \sum_{m=k}^{\infty}{}^{(m-k\ \text{even})} \frac{m!\Gamma(\frac{1}{2}m - \frac{1}{2}k + \frac{1}{2})}{(m-k)!\Gamma(\frac{1}{2}m + \frac{1}{2}k + \sigma + 1)} \psi_m(\mu, 0) \qquad \sigma \neq 0, \tag{24}$$

where the $\psi_m(\mu, 0)$ are the initial transformed matrix elements of the density operator.

The Gegenbauer polynomials have the following properties (see § 10.9 of Erdélyi *et al* 1953)

$$(\partial/\partial y)^l C_k^\sigma(y) = \begin{cases} [2^l \Gamma(\sigma + l)/\Gamma(\sigma)] C_{k-l}^{\sigma+l}(y) & \text{for } k \geqslant l \\ 0 & \text{for } k < l \end{cases} \tag{25}$$

and

$$C_k^\sigma(0) = \begin{cases} (-1)^{\frac{1}{2}k} \Gamma(\sigma + \frac{1}{2}k)/\Gamma(\sigma)\Gamma(1 + \frac{1}{2}k) & \text{for } k \text{ even} \\ 0 & \text{for } k \text{ odd.} \end{cases} \tag{26}$$

It therefore follows from (20), (21), (25) and (26) that

$$\psi_n(\mu, \tau) = \sum_{k=n}^{\infty}{}^{(k-n\ \text{even})} \frac{(-1)^{\frac{1}{2}k - \frac{1}{2}n} 2^n \Gamma(\frac{1}{2}k + \frac{1}{2}n + \sigma)}{n!\Gamma(\sigma)\Gamma(\frac{1}{2}k - \frac{1}{2}n + 1)} A_k^\sigma \exp(-\lambda_k \tau) \qquad \sigma \neq 0. \tag{27}$$

This result gives the general solution for the time dependence of the transformed matrix elements except that it excludes the case $\sigma = 0$ or $\mu = 1$. This exclusion arises from a $\sigma \neq 0$ restriction on an orthogonality relation used in the derivation of (24). This difficulty can be avoided by working instead with the Tchebichef polynomial of the first kind $T_k(y)$ related to the $\sigma = 0$ Gegenbauer polynomial by

$$C_k^0(y) = (1/2k) T_k(y). \tag{28}$$

The calculation now proceeds in the same way as that described earlier in the section, making use of the analogous integral and differential properties of the Tchebichef polynomials given for example in § 10.11 of Erdélyi *et al* (1953). The final results are

$$\psi_n(1, \tau) = \sum_{k=n}^{\infty}{}^{(k-n\ \text{even})} \frac{(-1)^{\frac{1}{2}k - \frac{1}{2}n} 2^{n-1} k \Gamma(\frac{1}{2}k + \frac{1}{2}n)}{n!\Gamma(\frac{1}{2}k - \frac{1}{2}n + 1)} B_k \exp(-k^2 \tau) \tag{29}$$

where

$$B_k = \sum_{m=k}^{\infty \,(m-k \text{ even})} [m!/2^{m-\delta(k)}(\tfrac{1}{2}m+\tfrac{1}{2}k)!(\tfrac{1}{2}m-\tfrac{1}{2}k)!]\psi_m(1,0) \tag{30}$$

and

$$\delta(k) = \begin{cases} 1 & \text{for } k>0 \\ 0 & \text{for } k=0. \end{cases} \tag{31}$$

Equations (24), (27), (29) and (30) provide complete solutions for the time dependences of all matrix elements of the density operator for all initial states of the light beam. The diagonal result obtained by setting $\mu=0$ and $\sigma=-\frac{1}{2}$ in (27) reproduces equation (63) in Simaan and Loudon (1975). In the steady state achieved by letting τ tend to infinity, it is clear from (23) that only matrix elements with $\mu=0$ or 1 can survive; in the latter case (29) is zero except for $n=0$, when it reduces to B_0, and (30) reproduces the result (11) for the only non-zero off-diagonal matrix element.

5. Discussion

The diagonal matrix elements of the density operator are fully discussed in our earlier paper and we make no further comment here except to show in figure 1 their time dependence for an initially coherent beam of light. The light in this case is antibunched for all times $\tau>0$ and the graph shows the way in which the diagonal distribution $\rho_{n,n}$ tends to its steady-state form with only $\rho_{0,0}$ and $\rho_{1,1}$ non-zero. Initially chaotic light shows a qualitatively similar behaviour for times τ greater than about 0·2 where it also becomes antibunched.

The properties of the off-diagonal matrix elements are best illustrated by evaluating their impact on some observable quantity. The electric-field expectation value of the light, depending on the $\mu=1$ matrix elements is a convenient quantity to consider. According to chapter 7 of Loudon (1973), the electric-field expectation value shows a

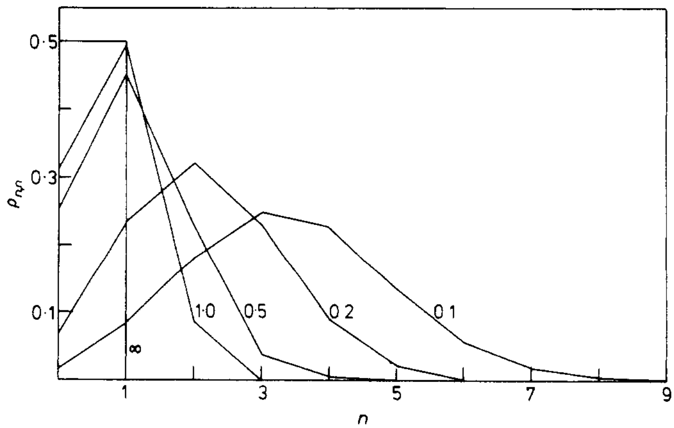

Figure 1. Diagonal distributions $\rho_{n,n}$ as functions of n for the times τ indicated against the curves for an initially coherent light beam with mean photon number equal to ten.

sine-wave behaviour with amplitude

$$E = (2\hbar\omega/\epsilon_0 V)^{1/2} \sum_n (n+1)^{1/2} |\rho_{n,n+1}|, \tag{32}$$

where V is the quantisation volume. For an initially coherent state specified by (8) and (12), the phase of the sine wave is θ at all times, and the value of the summation in (32) changes from $|\alpha|$ at $\tau = 0$ to $(2\pi)^{-1/2}$ at $\tau = \infty$ (from (17) assuming $|\alpha|^2 \gg 1$). Figure 2 shows the complete time dependence of the summation, calculated with the help of (29) and (30). The uncertainty ΔE in the electric field can also be calculated; it increases slightly from its constant value $(\hbar\omega/2\epsilon_0 V)^{1/2}$ in the initial coherent state to become comparable with the electric-field amplitude (32) in the steady state.

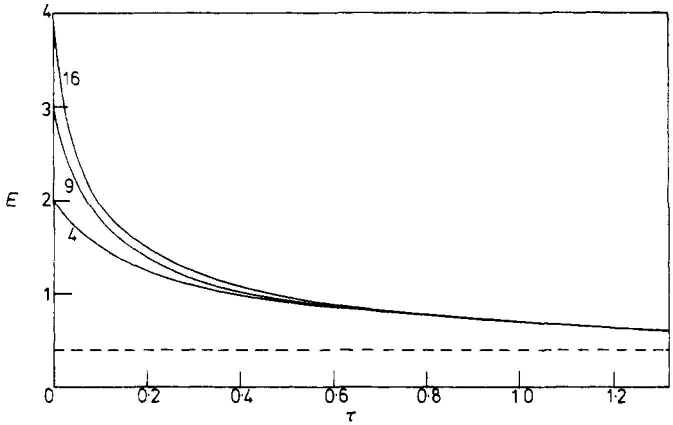

Figure 2. Time dependence of the electric-field amplitude for initially coherent light with the mean photon numbers shown on the curves. The quantity plotted on the vertical axis is the summation in (32) and the broken line is the steady-state asymptote at $(2\pi)^{-1/2}$.

The feasibility of making measurements of electric fields within the constraints imposed by quantum mechanics is very carefully considered by Bohr and Rosenfeld (1933). They conclude that such measurements are in principle possible provided that in deriving quantum-mechanical predictions, proper account is taken of the finite spatial and temporal extents necessarily present in practicable measurements. Such account has not been taken here and electric-field measurements at optical frequencies are not in any case currently possible. However, the results derived provide the basic information needed for more realistic calculations of the measurable electric field and more generally for other off-diagonal properties of two-photon absorbed light.

The electric-field variation explicitly derived here, that of a sine wave of fixed phase, diminishing amplitude and increasing uncertainty spread, adds to the known features of the initially-coherent antibunched light. The electric-field behaviour does not in fact show such remarkable effects as other features of antibunched light, particularly the predicted negative Hanbury Brown and Twiss correlation. Finally, it should be emphasised that in the example of initially coherent light, the light at all subsequent times cannot be described in classical terms, and this is particularly apparent in the steady state where the quantum-mechanical uncertainty ΔE is comparable to the field amplitude E.

Acknowledgment

We thank Eugene Philips for translating the paper by Bohr and Rosenfeld (1933) into English.

References

Bohr N and Rosenfeld L 1933 *K. Danske Vidensk. Selsk. Math.-Fys. Meddr* **12** No. 8 1–65
Chandra N and Prakash H 1970 *Phys. Rev.* A **1** 1696–8
Erdélyi A, Magnus W, Oberhettinger F and Tricomi F G 1953 *Higher Transcendental Functions* (New York: McGraw-Hill)
Loudon R 1973 *The Quantum Theory of Light* (Oxford: Clarendon)
—— 1976 *Phys. Bull.* **27** 21–3
McNeil K J and Walls D F 1974 *J. Phys. A: Math., Nucl. Gen.* **7** 617–31
Shen Y R 1967 *Phys. Rev.* **155** 921–31
Simaan H D and Loudon R 1975 *J. Phys. A: Math. Gen.* **8** 539–54