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Competition between one- and two-photon absorption processes

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Abstract. We obtain an exact analytical solution to the master equation for the diagonal density matrix elements of the one-mode quantized field, when both one- and two-photon absorption processes are present. Explicit expressions for the time dependences of the factorial moments are found. The special cases of the initial Fock's, binomial, negative binomial, thermal, and coherent states, as well as of their even/odd counterparts are considered in detail. The existence of the universal time-dependent distribution of initially highly excited states is discovered, and simple explicit expressions are given for some specific values of parameters. This distribution holds for times exceeding the transition time of the order of $(D_2\bar{n}_0)^{-1}$, D_2, $\bar{n}_0$ being the two-photon absorption coefficient and the initial mean photon number, respectively. The transition time from any initial state to the ground state is shown to be finite even for highly excited states, provided that $D_2 \neq 0$. Although the final stage of evolution is characterized by the sub-Poissonian statistics for any initial state, Mandel's parameter is shown to be very sensitive to small differences in high-order initial factorial moments at the intermediate stage.

1. Introduction

The process of quantum relaxation is described usually in the framework of the master equation for the density operator $\hat{\rho}$, which has the following structure (in the interaction picture) [1–3]:

$$\frac{\partial \hat{\rho}}{\partial t} = \frac{1}{2}\sum_n D_n(2\hat{A}_n\hat{\rho}\hat{A}_n^\dagger - \hat{A}_n^\dagger\hat{A}_n\hat{\rho} - \hat{\rho}\hat{A}_n^\dagger\hat{A}_n) \qquad D_n \geqslant 0 \tag{1}$$

where $\hat{A}_n$ may be an arbitrary linear operator. If the system under study is an electromagnetic field mode (or an equivalent harmonic oscillator), then each operator $\hat{A}_n$ can be expressed in terms of the annihilation and creation operators $\hat{a}$, $\hat{a}^\dagger$ satisfying the commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$. For instance, the choice $n = 2$, $\hat{A}_1 = \hat{a}$, $\hat{A}_2 = \hat{a}^2$ means that both one- and two-photon absorption processes are present. A consequence of equation (1) is the following set of equations for the occupation probabilities in the Fock basis, $p_n(t) = \langle n|\hat{\rho}(t)|n\rangle$,

$$\mathrm{d}p_n/\mathrm{d}t = D_1[(n+1)p_{n+1} - np_n] + D_2[(n+1)(n+2)p_{n+2} - n(n-1)p_n]. \tag{2}$$

Introducing the *generating function*

$$F(z,t) = \sum_{n=0}^{\infty} p_n(t)z^n \tag{3}$$

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one can replace this infinite system of coupled ordinary differential equations by a single partial differential equation

$$\partial F/\partial t = D_1(1-z)\partial F/\partial z + D_2(1-z^2\partial^2 F/\partial z^2. \qquad (4)$$

A general solution to this equation in the special case $D_2 = 0$ was found in [4, 5]:

$$F(z,t) = F_0(1+(z-1)\mathrm{e}^{-D_1 t}) \qquad F_0(z) \equiv F(z,0). \qquad (5)$$

A solution of equation (4) with $D_1 = 0$ was given in [6–10]:

$$F(z,t) = \sum_{n=0}^{\infty} b_n C_n^{(-1/2)}(z) \exp[-D_2 n(n-1)t] \qquad (6)$$

where $C_n^{(\alpha)}(z)$ is the Gegenbauer polynomial, and the coefficients b_n read

$$b_n = (n - \tfrac{1}{2}) \int_{-1}^{1} [F_0(z) - b_+ - b_- z] C_{n-2}^{(3/2)}(z)\,\mathrm{d}z \qquad n \geqslant 2 \qquad (7)$$

$$b_0 = b_+ \qquad b_1 = -b_- \qquad b_\pm = \tfrac{1}{2}[1 \pm F_0(-1)]. \qquad (8)$$

The results of the papers cited (for other references see, e.g. [11, 12]) show that one- and two-photon absorption processes are qualitatively different in many respects. For instance, the one-photon processes preserve the type of the photon statistics (Poissonian, sub- or super-Poissonian), whereas the two-photon absorption results in the antibunching effect, or sub-Poissonian statistics for any initial states. Due to this reason, it seems interesting to consider the case when both the processes, one- and two-photon, are present simultaneously, and to analyse the effects of their competition. A solution of equation (4) with $D_1 \neq 0$ and $D_2 \neq 0$ was expressed in terms of the Jacobi polynomials in a short article [13]. The aim of our paper is to provide a detailed investigation of the problem. In particular, we consider a large family of initial states, for which the explicit expressions for the coefficients in expansions like (6) can be found, and analyse different asymptotical and limit cases. We confine ourselves to finding analytical expressions for the time-dependent *diagonal* matrix elements of the density operator. The behaviour of the off-diagonal elements will be the subject of another publication.

The paper is organized as follows. In the next section we obtain a general solution to the master equation and demonstrate the existence of a universal distribution function in the case of highly excited initial states. In section 3 we consider the mean transition time to the ground state, and show that it is limited even for highly excited states, provided that the two-photon absorption coefficient is not equal to zero. In section 4 we give explicit expressions for the factorial moments and Mandel's $\mathcal{Q}$-factor in the general case. Different important special cases are considered in section 5. A summary and conclusion are given in section 6. The details of calculations are discussed in the appendix.

2. General solution to the master equation

It is natural to solve equation (4) by separation of variables: $F(z,t) = f(z)\mathrm{e}^{-\lambda t}$. Then function $f(z)$ must satisfy the equation

$$D_2(1-z^2)f'' + D_1(1-z)f' + \lambda f = 0.$$

Comparing it with the equation for the Jacobi polynomial $P_n^{(\alpha,\beta)}(x)$ [14, 15]

$$(1-x^2)f'' + [\beta - \alpha - (\beta+\alpha+2)x]f' + n(n+\beta+\alpha+1)f = 0$$

we obtain the following general solution to equation (4) (see also [13]):

$$F(z,t) = 1 + \sum_{n=1}^{\infty} A_n P_n^{(-1,\nu-1)}(z) \mathrm{e}^{-\lambda_n t} \tag{9}$$

$$\nu = D_1/D_2 \qquad \lambda_n = D_1 n + D_2 n(n-1) = D_2 n(n+\nu-1). \tag{10}$$

The explicit form of the polynomial $P_n^{(-1,\beta)}(z)$ with $n \geqslant 1$ is (hereafter $\beta = \nu - 1$)

$$P_n^{(-1,\beta)}(z) = \frac{1}{n} \sum_{k=1}^{n} \frac{(-n)_k (\beta+n)_k}{k!(k-1)!} \left(\frac{1-z}{2}\right)^k = (n-1)! \sum_{k=1}^{n} \frac{(\beta+n)_k}{k!(k-1)!(n-k)!} \left(\frac{z-1}{2}\right)^k \tag{11}$$

so the normalization condition $F(1,t) \equiv 1$ is fulfilled. The coefficients A_n must be determined from the initial function $F_0(z)$. The delicate point is that the Jacobi polynomials $P_n^{(\alpha,\beta)}(x)$ form a complete orthonormal set in the interval $(-1, 1)$ with the weight factor $(x-1)^\alpha (x+1)^\beta$ under the condition $\alpha > -1$, $\beta > -1$. In the case under study $\alpha = -1$. None the less, one can check that for $m, n \geqslant 1$ the following relations hold (see the appendix):

$$\int_{-1}^{1} (1-x)^{-1} (1+x)^\beta P_n^{(-1,\beta)}(x) P_m^{(-1,\beta)}(x)\, \mathrm{d}x = \begin{cases} \dfrac{2^\beta (n+\beta)}{n(2n+\beta)} & m = n \\ 0 & m \neq n. \end{cases} \tag{12}$$

Thus, the coefficients A_n can be calculated as follows:

$$A_n = 2^{-\beta} \frac{n(2n+\beta)}{(n+\beta)} \int_{-1}^{1} [F_0(z) - 1](1-z)^{-1}(1+z)^\beta P_n^{(-1,\beta)}(z)\, \mathrm{d}z. \tag{13}$$

The integral on the right-hand side exists because $P_n^{(-1,\beta)}(1) = 0$. Moreover, using Rodrigues' formula for the Jacobi polynomials and integration by parts, we can transform (13) into the form (if $\nu > 0$)

$$A_n = 2^{-\nu} \frac{(2n+\beta)}{(n+\beta)} \int_{-1}^{1} F_0'(z)(1+z)^\nu P_{n-1}^{(0,\nu)}(z)\, \mathrm{d}z \tag{14}$$

whose integrand is free of singularities. At $\nu = 0$ solution (9) goes into (6) with $b_n = A_n(1-n)/2$ (for $n \geqslant 2$). Due to equations (3), (9), and the relation [14, 15]

$$(\mathrm{d}/\mathrm{d}x)^m P_n^{(\alpha,\beta)}(x) = 2^{-m}(n+\alpha+\beta+1)_m P_{n-m}^{(\alpha+m,\beta+m)}(x) \tag{15}$$

the time-dependent occupation probabilities can be expressed as

$$p_m(t) = \sum_{n=m}^{\infty} A_n B_{nm}^{(\nu)} \mathrm{e}^{-\lambda_n t} \tag{16}$$

with

$$\begin{aligned} B_{nm}^{(\nu)} &= \frac{(n+\nu-1)_m}{2^m m!} P_{n-m}^{(m-1,m+\nu-1)}(0) \\ &= \frac{(-1)^{n-m}(n+\nu-1)_m (m+\nu)_{n-m}}{2^n m!(n-m)!} F(m-n, 1-n; m+\nu; -1). \end{aligned} \tag{17}$$

$F(a, b; c; z)$ is the Gauss hypergeometric function. In particular, the occupation probabilities of the ground and the first-excited states read

$$p_0(t) = 1 + \sum_{n=1}^{\infty} A_n \frac{(-1)^n (\nu)_n}{2^n n!} F(-n, 1-n; \nu; -1) e^{-\lambda_n t} \tag{18}$$

$$p_1(t) = \sum_{n=1}^{\infty} A_n \frac{(-1)^{n-1} (1+\nu)_{n-1} (n+\nu-1)}{2^n (n-1)!} F(1-n, 1-n; 1+\nu; -1) e^{-\lambda_n t}. \tag{19}$$

For $D_2 = 0$, solution (5) can be written as follows,

$$F(z,t) = \sum_{k=0}^{\infty} p_k(0)[1 + (z-1)e^{-D_1 t}]^k$$
$$= \sum_{k=0}^{\infty} p_k(0) \sum_{m=0}^{k} (z-1)^m \binom{k}{m} e^{-D_1 t m} = \sum_{m=0}^{\infty} (z-1)^m e^{-D_1 t m} \sum_{k=m}^{\infty} p_k(0) \binom{k}{m}.$$

If $0 < D_2 \ll D_1$, then $\beta \gg 1$, and formula (11) can be simplified:

$$P_n^{(-1,\beta)}(z) \approx \frac{(\beta y)^n}{n!}[1 + \mathcal{O}(n^2/\beta y)] \qquad \beta \gg 1 \qquad y = \frac{z-1}{2}.$$

Evidently, for $\nu \gg 1$ and sufficiently large values of t, the small corrections of the order of $\mathcal{O}(1/\nu)$ in the pre-exponential coefficients of expansion (9) are not significant, so one should take into account the changes in the arguments of the exponential functions only. Then we have an approximate formula

$$F(z,t) \approx \sum_{m=0}^{\infty} \frac{\mathcal{N}_m(0)}{m!} (z-1)^m e^{-\lambda_m t} \tag{20}$$

where

$$\mathcal{N}_m \equiv \sum_{n=m}^{\infty} n(n-1)\dots(n-m+1) p_n = \partial^m F / \partial z^m |_{z=1} \tag{21}$$

is the mth factorial moment. In this approximation,

$$p_n(t) \approx \frac{1}{n!} \sum_{m=n}^{\infty} \frac{\mathcal{N}_m(0)}{(m-n)!} (-1)^{m-n} e^{-\lambda_m t}$$

so the occupation probability of the mth level at $t \gg 1/D_1$ is determined by the initial mth factorial moment: $p_m(t \gg 1/D_1) \approx (\mathcal{N}_m(0)/m!) \exp(-\lambda_m t)$. However, this is true provided that the initial mean photon number is not too large, $\bar{n}_0 \equiv \mathcal{N}_1(0) \ll \nu$.

In the opposite case, $\bar{n}_0 \gg \nu$, the final stage of the evolution turns out to be *independent of the initial conditions*. Indeed, for large values of $\bar{n}_0$, the function $F_0'(z)$ has a sharp maximum at $z = 1$, $F_0'(1) = \bar{n}_0$, whose width does not exceed $\delta z \sim 1/\bar{n}_0$ (because $F_0(z) \geqslant 0$ for $z \geqslant 0$). Therefore the main contribution to integral (14) is given by the interval $(1-\delta z, 1)$ (in general, function $F_0'(z)$ may also possess a large maximum or minimum at point $z = -1$, but its contribution is supressed by the factor $(1+z)^\nu$ in the integrand). If $n \ll \bar{n}_0$, then function $P_{n-1}^{(0,\nu)}(z)$ almost does not vary in the interval $(1-\delta z, 1)$, so we may replace it by $P_{n-1}^{(0,\nu)}(1) \equiv 1$ and write $(1+z)^\nu \approx 2^\nu$. Thus, we arrive at the asymptotical formula (see also [13])

$$A_n^{(\infty)} = \frac{2n-1+\nu}{n-1+\nu} \qquad n \ll \bar{n}_0. \tag{22}$$

Consequently, in *any state* satisfying the initial condition $\bar{n}_0 \gg \max(\nu, 1)$, the evolution of the occupation probabilities is described, after a short transient time of the order of $(D_2\bar{n}_0)^{-1}$ (see the end of section 4), by the universal formula

$$p_m^{(\infty)}(t) = \sum_{n=m}^{\infty} G_{nm}^{(\nu)} \mathrm{e}^{-\lambda_n t} \tag{23}$$

$$G_{nm}^{(\nu)} = \frac{(-1)^{n-m}(2n-1+\nu)(n+\nu)_{m-1}(m+\nu)_{n-m}}{2^n m!(n-m)!} F(m-n, 1-n; m+\nu; -1). \tag{24}$$

If ν is an integer, then $F(m-n, 1-n; m+\nu; -1)$ can be expressed as a finite combination of gamma-functions (see the appendix), and simpler formulae for $B_{nm}^{(\nu)}$ and $G_{nm}^{(\nu)}$ can be found. For example, for $\nu = 1$ we obtain $A_n^{(\infty)} \equiv 2$ for all n, so $G_{nm}^{(1)} = 2B_{nm}^{(1)}$. Then, using equation (A.2), we obtain

$$G_{m+2k,m}^{(1)} = -G_{m+2k+1,m}^{(1)} = \frac{(-1)^k 2^m (\frac{1}{2})_{m+k}}{m!k!} \qquad \lambda_n = n^2. \tag{25}$$

Figure 1 shows the evolution of the photon number distribution in this case. Also, it shows a perfect coincidence with the Gaussian interpolation formula

$$p(n,t) = [4\pi\bar{n}(t)/3]^{-1/2} \exp\left[-\frac{3(n-\bar{n}(t))^2}{4\bar{n}(t)}\right] \tag{26}$$

proposed (for *pure two-photon* processes) in [16]. The sub-Poissonian distribution (26) possesses the dispersion $\sigma_n = \frac{2}{3}\bar{n}$, in agreement with equation (52) at $D_1 t \ll 1$. With the increase of ν, the curves are shifted to the left without significant changes to their shapes.

The case $\nu = 0$ (pure two-photon absorption) needs a special analysis, because the subsets consisting of even and odd Fock's states do not mix, so that we have two additional constants of motion, b_+ and b_- (8), and two universal distributions for highly excited initial

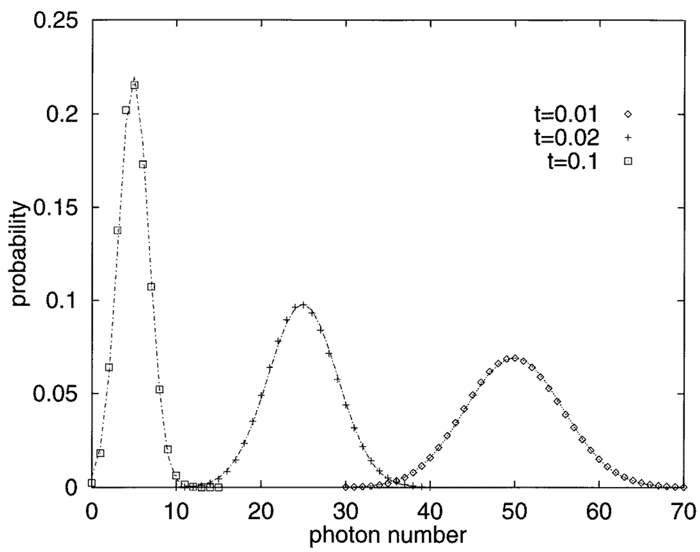

Figure 1. The asymptotical photon number distributions for $D_1 = D_2 = 1$ and $t = 0.01$ (right peak), $t = 0.02$ (middle peak), and $t = 0.1$ (left peak). The continuous curves represent the interpolation formula (26) with the same mean photon numbers.

states, respectively. In this case equation (16) reads (we use equation (A.1))

$$p_m(t,\nu=0)=\sum_{k=0}^{\infty}A_{m+2k}\frac{(-1)^k(m+2k-1)2^m(\frac{1}{2})_{m+k-1}}{4m!k!}\mathrm{e}^{-\lambda_{m+2k}t}\qquad m\geqslant 2 \tag{27}$$

$$p_1(t,\nu=0)=b_-+\sum_{k=1}^{\infty}A_{1+2k}\frac{(-1)^k(\frac{1}{2})_k}{(k-1)!}\mathrm{e}^{-2k(2k+1)D_2t} \tag{28}$$

$$p_0(t,\nu=0)=b_++\sum_{k=1}^{\infty}A_{2k}\frac{(-1)^k(\frac{1}{2})_k}{2k!}\mathrm{e}^{-2k(2k-1)D_2t}. \tag{29}$$

Evaluating the coefficients A_n for highly excited initial states, we cannot neglect now the contribution of the domain near $z=-1$. But since the Legendre polynomial $P_{n-1}^{(0,0)}(z)$ possesses the definite parity $(-1)^{n-1}$, only even or odd parts of function $F_0'(z)$ yield the contribution to the integral. Thus the domain of integration can be reduced to the interval $(0,1)$. Then, using the same reasonings as above, we may replace $P_{n-1}^{(0,0)}(z)$ by $P_{n-1}^{(0,0)}(1)=1$. Assuming that the occupation probabilities of the lowest levels ($p_0, p_1, \ldots$) go to zero when $\bar{n}_0\to\infty$ (this is the usual case in the physical applications), we arrive at the following universal formulae for the evolution of highly excited initial states due to the pure two-photon absorption:

$$p_{2l}^{(\infty)}(t,\nu=0)=b_+\sum_{k=0}^{\infty}\frac{(-1)^k(2l+2k-\frac{1}{2})2^{2l}(\frac{1}{2})_{2l+k-1}}{(2l)!k!}\mathrm{e}^{-\lambda_{2l+2k}t}\qquad l\geqslant 1 \tag{30}$$

$$p_{2l+1}^{(\infty)}(t,\nu=0)=b_-\sum_{k=0}^{\infty}\frac{(-1)^k(4l+4k+1)2^{2l}(\frac{1}{2})_{2l+k}}{(2l+1)!k!}\mathrm{e}^{-\lambda_{2l+2k+1}t}\qquad l\geqslant 1 \tag{31}$$

$$p_1^{(\infty)}(t,\nu=0)=b_-\left[1+\sum_{k=1}^{\infty}\frac{(-1)^k(4k+1)(\frac{1}{2})_k}{k!}\mathrm{e}^{-2k(2k+1)D_2t}\right] \tag{32}$$

$$p_0^{(\infty)}(t,\nu=0)=b_+\left[1+\sum_{k=1}^{\infty}\frac{(-1)^k(4k-1)(\frac{1}{2})_{k-1}}{2k!}\mathrm{e}^{-2k(2k-1)D_2t}\right]. \tag{33}$$

In the special case $b_+=b_-=\frac{1}{2}$ these relations were found in [10].

For any nonzero value of ν, the mean value of the projection operator to the subspace of the Fock states with odd numbers of photons, $b_-(t)=\frac{1}{2}[1-F(-1,t)]$, eventually goes to zero. Due to equation (9),

$$b_-(t)=\frac{1}{2}\sum_{n=1}^{\infty}(-1)^{n-1}A_n\frac{(\nu)_n}{n!}\mathrm{e}^{-\lambda_nt}. \tag{34}$$

Two evident limiting cases result in the following simple expressions:

$$b_-(t\to\infty)=\frac{\nu}{2}A_1\mathrm{e}^{-D_1t}+\cdots\qquad b_-(t\to 0)=b_-(0)-D_1tF'(-1)+\cdots.$$

For highly excited initial states, $\bar{n}_0\gg\max(\nu,1)$, we obtain

$$b_-^{(\infty)}(t)=\frac{1}{2}\sum_{n=1}^{\infty}(-1)^{n-1}\frac{(2n+\nu-1)(\nu)_{n-1}}{n!}\mathrm{e}^{-\lambda_nt}. \tag{35}$$

In particular,

$$b_-^{(\infty)}(t,\nu\ll 1)=\frac{1}{2}\mathrm{e}^{-D_1t}+\nu\sum_{n=2}^{\infty}(-1)^{n-1}\frac{(2n-1)}{2n(n-1)}\mathrm{e}^{-\lambda_nt}.$$

This example shows that the limit transition $\nu\to 0$ is not uniform, and the case $\nu=0$ is peculiar.

3. Mean decay time

The difference between the cases $\nu = 0$ and $\nu \ll 1$ is seen distinctly when one analyses the *transition time* to the ground state. Different possible definitions of the transition times have already been discussed in [17], as well as numerous examples for pure one- or two-photon processes. Here we confine ourselves to the *mean transition time*

$$\overline{T} = [1 - p_0(0)]^{-1} \int_0^\infty t(\mathrm{d}p_0/\mathrm{d}t)\,\mathrm{d}t = [1 - p_0(0)]^{-1} \int_0^\infty [1 - p_0(t)]\,\mathrm{d}t. \tag{36}$$

This definition can be justified by the observation that the function $g(t) = [1 - p_0(0)]^{-1}\,\mathrm{d}p_0/\mathrm{d}t$ satisfies the relations $g(t) \geqslant 0$, $\int_0^\infty g(t)\,\mathrm{d}t = 1$, so $g(t)\,\mathrm{d}t$ can be considered as the probability of reaching the ground state at the time interval between t and $t + \mathrm{d}t$. Using equation (18) we obtain

$$\begin{aligned}\overline{T} &= \frac{\nu}{D_1[1 - p_0(0)]} \sum_{n=1}^{\infty} A_n \frac{(-1)^{n-1}(\nu)_{n-1}}{n 2^n n!} F(-n, 1-n; \nu; -1) \\ &= \frac{\nu}{D_1[1 - p_0(0)]} \left[\frac{A_1}{2} - \sum_{n=2}^{\infty} \frac{A_n}{n(n+\nu-1)} P_n^{(-1,\nu-1)}(0) \right].\end{aligned} \tag{37}$$

If $\bar{n}_0 \to \infty$, we may replace the coefficient A_n with its asymptotical form (22). Consequently, the transition time is finite even for highly excited initial states and for any fixed values of D_2 and ν:

$$\begin{aligned}\overline{T}^{(\infty)}(\nu) &= \frac{\nu}{D_1} \sum_{n=1}^{\infty} \frac{(-1)^{n-1}(\nu)_{n-1}(2n+\nu-1)}{n 2^n n!(n+\nu-1)} F(-n, 1-n; \nu; -1) \\ &= \frac{1+\nu}{2D_1} - \frac{\nu}{D_1} \sum_{n=2}^{\infty} \frac{2n+\nu-1}{n(n+\nu-1)^2} P_n^{(-1,\nu-1)}(0)\end{aligned} \tag{38}$$

(for the sake of simplicity, we assume that $p_0(0) \to 0$ when $\bar{n}_0 \to \infty$). Since the value of the Jacobi polynomial $P_n^{(-1,\nu-1)}(0)$ remains finite at $\nu \to 0$ for $n \geqslant 2$ (see (11)), we have $\lim_{\nu\to 0} \overline{T}^{(\infty)}(\nu) = 1/(2D_1)$, whereas for the pure two-photon absorption [17] $\overline{T}^{(\infty)}(D_1 = 0) = D_2^{-1} \ln 2$. The origin of the difference is quite clear: for small, but nonzero values of ν, the first-excited Fock state $|1\rangle$ is metastable, and its decay time is proportional to D_1^{-1}. The plot of function $\overline{T}^{(\infty)}(\nu)$ is given in figure 2.

If $\nu \gg 1$, the value of the hypergeometric function in (38) becomes close to unity, and $(\nu)_k \approx \nu^k$. In this limiting case we obtain

$$\overline{T}^{(\infty)}(\nu \gg 1) \approx -D_1^{-1} \sum_{n=1}^{\infty} \frac{(-\nu/2)^n}{n n!} = D_1^{-1}[\ln(\nu/2) + \mathrm{C} - \mathrm{Ei}(-\nu/2)] \tag{39}$$

where $\mathrm{C} = 0.577\ldots$ is Euler's constant, and $\mathrm{Ei}(x)$ is the exponential-integral function [15]. Due to the asymptotical formula $\mathrm{Ei}(-x) \sim (-x)^{-1}\mathrm{e}^{-x}$ for $x \gg 1$, only two first terms inside the square brackets in (39) are significant at $\nu \gg 1$. If $D_2 = 0$, then the decay time depends on the initial state, and it goes to infinity with the increase of the initial mean photon number, approximately as $\ln(\bar{n}_0)$ [17]. Thus the case $\nu = \infty$ is also distinguished.

4. Mean values and factorial moments

Taking into account equations (9), (15), and the relations

$$P_n^{(0,\beta)}(1) = 1 \qquad P_n^{(1,\beta)}(1) = 1 + n \qquad P_{n-m}^{(m-1,\beta+m)}(1) = \binom{n-1}{n-m}$$

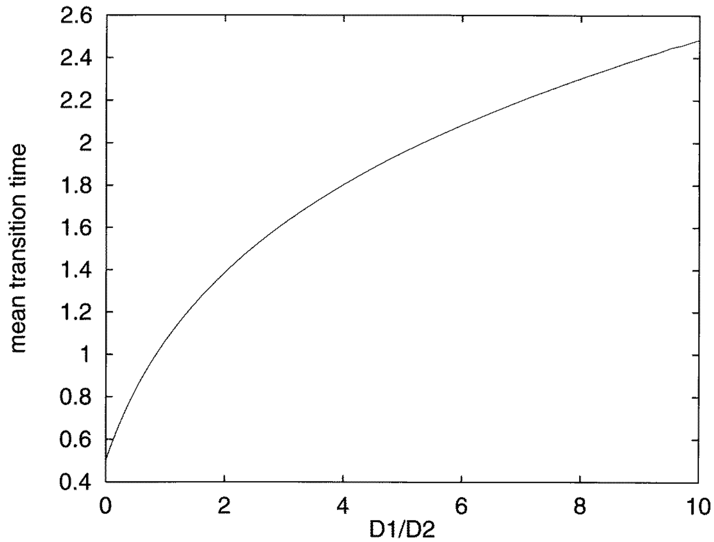

Figure 2. The mean transition time from the asymptotical state $\overline{T}^{(\infty)}$ (in the units of D_1^{-1}) versus $\nu = D_1/D_2$.

we obtain the following expressions for the factorial moments (21):

$$\mathcal{N}_m = \frac{1}{2^m(m-1)!}\sum_{n=m}^{\infty}(n-m+1)_{m-1}(n+\nu-1)_m A_n \mathrm{e}^{-\lambda_n t}. \tag{40}$$

In particular,

$$\overline{n} \equiv \mathcal{N}_1 = \tfrac{1}{2}\sum_{n=1}^{\infty}(n+\nu-1)A_n \mathrm{e}^{-\lambda_n t} \tag{41}$$

$$\overline{n(n-1)} \equiv \mathcal{N}_2 = \tfrac{1}{4}\sum_{n=2}^{\infty}(n-1)(n+\nu)(n+\nu-1)A_n \mathrm{e}^{-\lambda_n t}. \tag{42}$$

For $D_1 t \gg 1$ or $D_2 t \gg 1$, it is sufficient to retain only the first terms of the series. In the framework of approximation (20) (i.e. for $D_1 \gg D_2$) we obtain

$$\mathcal{N}_m(t) \approx \mathcal{N}_m(0)\mathrm{e}^{-\lambda_m t}. \tag{43}$$

Consequently, Mandel's parameter

$$\mathcal{Q} = [\overline{n(n-1)} - (\overline{n})^2]/\overline{n} \tag{44}$$

depends on time as

$$\mathcal{Q}(t) = \mathrm{e}^{-D_1 t}[\mathcal{Q}(0)\mathrm{e}^{-2D_2 t} - \overline{n}_0(1-\mathrm{e}^{-2D_2 t})]. \tag{45}$$

When $t \to \infty$, $\mathcal{Q}(t)$ becomes negative (the signature of the sub-Poissonian statistics, which is considered usually as evidence of the quantum nature of light) *for any initial value* $\mathcal{Q}(0)$, provided that $D_2 > 0$. The type of the photon statistics (Poissonian, sub- or super-Poissonian) is not changed in the one-photon absorption processes only, whereas even a small admixture of the two-photon absorption will always result ultimately in a sub-Poisson statistics, provided that time, t, is sufficiently large. The reason is clear: in the presence of the two-photon absorption, the second factorial moment decreases faster than the square of the mean photon number.

For highly excited initial states, according to equation (22), we have the universal formulae ($\nu > 0$)

$$\overline{n}^{(\infty)}(t) = \tfrac{1}{2}\sum_{n=1}^{\infty}(2n+\nu-1)\mathrm{e}^{-\lambda_n t} \tag{46}$$

$$\overline{n(n-1)}^{(\infty)}(t) = \tfrac{1}{4}\sum_{n=2}^{\infty}(n-1)(n+\nu)(2n+\nu-1)\mathrm{e}^{-\lambda_n t} \tag{47}$$

$$\mathcal{N}_m^{(\infty)}(t) = \sum_{n=m}^{\infty}\frac{(n-m+1)_{m-1}(n+\nu)_{m-1}(2n+\nu-1)}{2^m(m-1)!}\mathrm{e}^{-\lambda_n t}. \tag{48}$$

In particular, at the final stages of the evolution, when $D_1 t \gg 1$ or $D_2 t \gg 1$, only terms with $n = m$ are significant, and we obtain

$$\mathcal{N}_m^{(\infty)}(t) \approx 2^{-m}(m+\nu)_{m-1}(2m+\nu-1)\mathrm{e}^{-\lambda_m t}. \tag{49}$$

If $1 \ll \nu \ll \overline{n}_0$, then the pre-exponential factor in (49) equals simply $(D_1/2D_2)^m$. Mandel's parameter (44) turns out negative in the limiting case discussed. For example, if $\nu \gg 1$, then

$$\mathcal{Q}^{(\infty)}(t) \approx -\frac{\nu}{2}\mathrm{e}^{-D_1 t}[1-\mathrm{e}^{-2D_2 t}]$$

so that $\mathcal{Q}^{(\infty)}(t) \approx -D_1 t\mathrm{e}^{-D_1 t}$ in the time interval $D_1^{-1} \ll t \ll D_2^{-1}$. However, one cannot put $D_2 = 0$, since the formulae presented above hold for $\overline{n}_0 \gg D_1/D_2$. *Two-photon processes are necessary to cause a highly excited system to 'forget' its initial state*. In the presence of one-photon processes only, the system always 'remembers' its initial state, in accordance with equation (43), which becomes exact for $D_2 = 0$. If $\nu \ll \overline{n}_0$ and $D_2 t \gg 1$, then $\mathcal{Q}^{(\infty)}(t) \approx -\frac{1}{2}\mathrm{e}^{-D_1 t}$.

For small values of t, the number of terms yielding significant contributions to the right-hand sides of (46)–(48) becomes very large, whereas the magnitude of each summand is only slightly changed under the shift $n \to n+1$. Therefore, we may replace the summation by integration. It is remarkable that all the sums contain the term $(2n+\nu-1)$, which is proportional to the derivative $\mathrm{d}\lambda_n/\mathrm{d}n$. Consequently, the integration over n can be replaced by the integration over λ_n from 0 to ∞. Then the integrals can be calculated exactly, and we obtain

$$\mathcal{N}_1^{(\infty)}(t \to 0) = \frac{1}{2D_2 t} + \frac{1}{12}(2-3\nu) + \frac{D_2 t}{120}(4-10\nu+5\nu^2) + \cdots \tag{50}$$

$$\mathcal{N}_2^{(\infty)}(t \to 0) = \frac{1}{4D_2^2}\left(\frac{1}{t^2} - \frac{D_1}{t}\right) + \frac{1}{240}(25\nu^2-10\nu-4) + \cdots. \tag{51}$$

The nondivergent (at $t \to 0$) terms are obtained with the aid of the Euler–Maclaurin formula (A.3). They are necessary to obtain the correct expression for the $\mathcal{Q}$-factor up to the linear terms:

$$\mathcal{Q}^{(\infty)}(t \to 0) = -\frac{1}{3} + \left(\frac{D_1}{12} - \frac{2D_2}{45}\right)t + \cdots. \tag{52}$$

The plots of $\mathcal{Q}^{(\infty)}(t)$ for different values of ν are given in figure 3.

It is possible to find simple interpolation formulae describing the evolution of the factorial moments $\mathcal{N}_m(t)$ in the whole interval $0 < t < \infty$. From (4) and (21) we obtain the equations

$$\dot{\mathcal{N}}_1 = -D_1\mathcal{N}_1 - 2D_2\mathcal{N}_2 \tag{53}$$

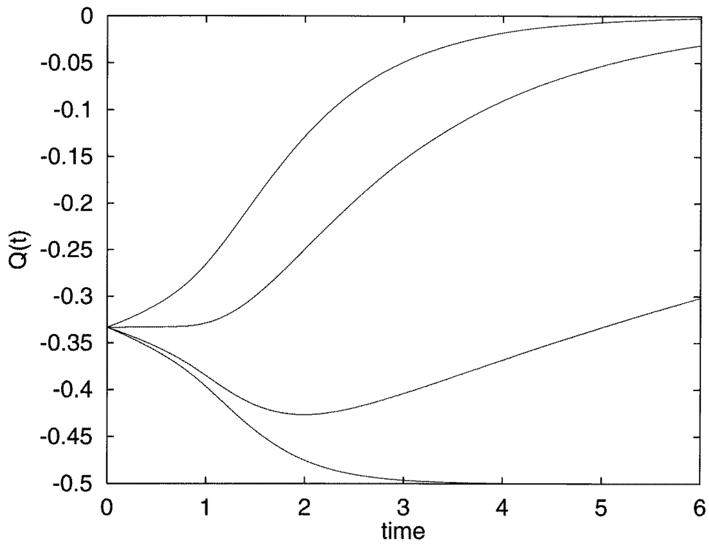

Figure 3. The time dependence of the $\mathcal{Q}$-factor in the asymptotical state. The curves from bottom to the top correspond to the following values of parameter $\nu = D_1/D_2$: $\nu = 0, 0.1, \frac{24}{45}, 1$. $D_2 = 1$.

$$\dot{\mathcal{N}}_2 = -2(D_1 + D_2)\mathcal{N}_2 - 4D_2\mathcal{N}_3 \tag{54}$$

$$\dot{\mathcal{N}}_m = -mD_1\mathcal{N}_m - D_2[2m\mathcal{N}_{m+1} + m(m-1)\mathcal{N}_m]. \tag{55}$$

To disentangle this infinite system of coupled equations, we make an assumption, that the factorial moments are related in the same manner as in the coherent states (which are considered usually as the 'most classical' states), i.e. $\mathcal{N}_m = \mathcal{N}_1^m$. Namely, making the substitution $\mathcal{N}_2 = \mathcal{N}_1^2$ in (53) and $\mathcal{N}_{m+1} = \mathcal{N}_m\mathcal{N}_1$ in (54), (55) we arrive at simplified (although approximate, of course) equations, which can be easily solved:

$$\mathcal{N}_m^{(\mathrm{clas})}(t) = \mathcal{N}_m(0)\mathrm{e}^{-\lambda_m t}[1 + 2(D_2/D_1)\mathcal{N}_1(0)(1 - \mathrm{e}^{-D_1 t})]^{-m}. \tag{56}$$

Other approximate solutions for the moments $\overline{n^l}$ were obtained in the special case of $D_1 = 0$ in [16].

If $t \to 0$ and $\mathcal{N}_1(0) \to \infty$, then the leading term of the expansion of the right-hand side of equation (56), $\mathcal{N}_m = (2D_2t)^{-m} + \cdots$, is the same (for $m = 1, 2$) as the leading terms in equations (50) and (51), provided that $\mathcal{N}_2(0) = \mathcal{N}_1^2(0)$ and $D_2\mathcal{N}_1(0)t \gg 1$. Thus, we obtain the applicability condition of the universal distributions for highly excited initial states: *all the formulae labelled by the superscript* $^{(\infty)}$ *hold for* $t \gg 1/(D_2\bar{n}_0)$.

In the next section we shall demonstrate that the coincidence of the exact and approximate solutions is quite good even for rather small initial values of the mean photon number: see figure 4. However, more delicate characteristics of the photon number distribution, such as the $\mathcal{Q}$-factor, are not described by simple interpolation formulae when $D_{1,2}t \sim 1$: see figure 5.

5. Examples

For the intermediate values of the initial parameters and time, one needs the concrete values of the coefficients A_n in the decomposition (9). Fortunately, the explicit analytical expressions for these coefficients can be found in many important special cases.

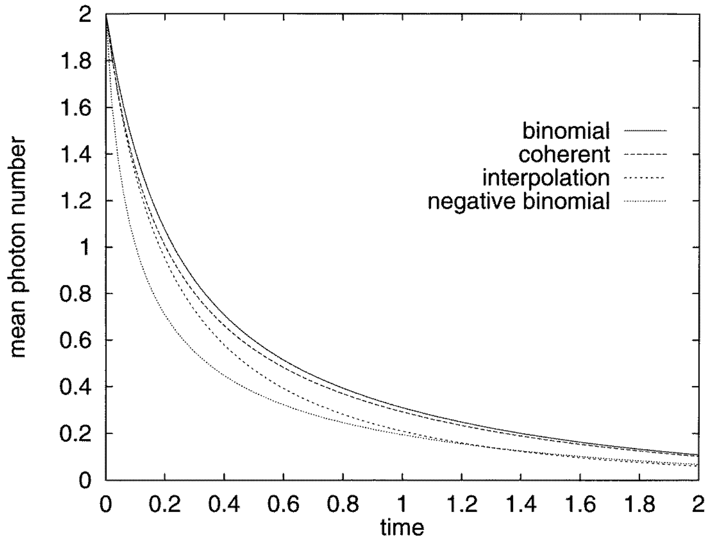

Figure 4. The time dependence of the mean photon number for different states with the same initial value $\bar{n}_0 = 2$. The order of the curves in the middle part of the plot (from top to bottom): binomial state with $M = 4, \eta = \frac{1}{2}$; coherent state with $a = 2$; interpolation formula (56); negative binomial states with $\mu = \frac{1}{2}$, $\xi = \frac{4}{5}$. $D_1 = D_2 = 1$.

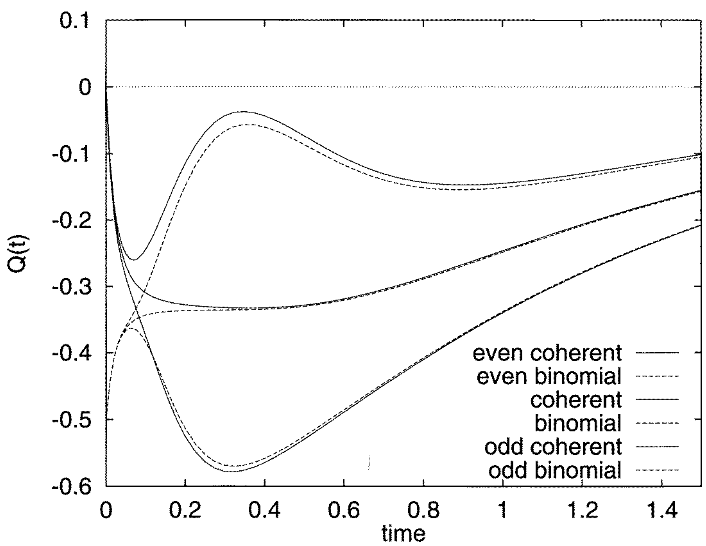

Figure 5. The time dependence of the $\mathcal{Q}$-factor for coherent and binomial initial states and for their even and odd counterparts. The two upper curves (at large values of time) correspond to even states, the two middle curves correspond to original coherent and binomial states, and the two lower curves correspond to odd states. The initial mean photon number equals $\bar{n}_0 = 4$ in all the cases. $\mathcal{Q}(0) = -0.5$ for all types of the binomial states, whereas $\mathcal{Q}(0)$ is close to zero (with opposite signs) for even/odd coherent states, and $\mathcal{Q}(0) = 0$ for coherent states. $D_1 = 1$, $\nu = 0.5$.

5.1. Evolution of initial coherent states

Let us begin with the initial *coherent* state $|\alpha\rangle$, which has the following properties ($a = |\alpha|^2$):

$$p_n(0) = \mathrm{e}^{-a}a^n/n! \qquad F_0(z) = \exp[a(z-1)] \qquad \bar{n}_0 = a \qquad \mathcal{N}_m = a^m \qquad \mathcal{Q} = 0.$$

Then we obtain (the details of the calculations are given in the appendix)

$$A_n^{(\mathrm{coh})} = (2a)^n \frac{\Gamma(n-1+\nu)}{\Gamma(2n-1+\nu)} \Phi(n; \nu+2n; -2a) \tag{57}$$

$\Phi(a; c; z)$ being the Kummer confluent hypergeometric function. At $\nu = 0$ function $\Phi(n; 2n; z)$ is reduced to the Bessel function, and we recover the result of [10],

$$b_n^{(\mathrm{coh})} = -(n - \tfrac{1}{2})\sqrt{2\pi a}\,\mathrm{e}^{-a} I_{n-1/2}(a).$$

If $D_2 \ll D_1$, then $\nu \gg 1$, so we can write $\Phi(n; \nu+2n; -2a) \approx 1 - 2an/\nu + \mathcal{O}((an/\nu)^2)$. Neglecting the terms of the order of $\mathcal{O}((an/\nu)^2)$, we obtain for the mean photon number

$$\bar{n}(t) = \bar{n}_0 \mathrm{e}^{-D_1 t} - \frac{2a^2}{\nu}\mathrm{e}^{-D_1 t}[1 - \mathrm{e}^{-D_1 t - 2D_2 t}]. \tag{58}$$

For Mandel's parameter we obtain the same expressions as given by equation (45), the corrections being of the order of a/ν:

$$\mathcal{Q}^{(\mathrm{coh})}(t) = -a\mathrm{e}^{-D_1 t}(1 - \mathrm{e}^{-2D_2 t}). \tag{59}$$

Using the known integral representation of the confluent hypergeometric function [15], we can rewrite (57) as ($n \geqslant 1$)

$$\begin{aligned} A_n^{(\mathrm{coh})} &= \frac{(2a)^n}{(n-1)!}\frac{(2n-1+\nu)}{(n-1+\nu)} \int_0^1 \mathrm{e}^{-2at} t^{n-1}(1-t)^{n+\nu-1}\,\mathrm{d}t \\ &= \frac{1}{(n-1)!}\frac{(2n-1+\nu)}{(n-1+\nu)} \int_0^{2a} \mathrm{e}^{-x} x^{n-1}\left(1 - \frac{x}{2a}\right)^{n+\nu-1}\mathrm{d}x. \end{aligned} \tag{60}$$

This form is convenient for the numerical calculations. Moreover, if $a \gg 1$, one can replace $[1 - x/(2a)]^{n+\nu-1}$ by $\exp[-(n+\nu-1)x/(2a)]$ and perform the integration over x from 0 to ∞. In this way we obtain the following asymptotics:

$$A_n^{(\mathrm{coh})} \approx \frac{(2n-1+\nu)}{(n-1+\nu)}\left(1 + \frac{\nu+n-1}{2a}\right)^{-n} \qquad a \gg 1.$$

If $a \gg 1$ and $\nu \gg 1$, then

$$A_n^{(\mathrm{coh})} \approx \left(\frac{2a}{2a+\nu}\right)^n \tag{61}$$

so at $D_1 t \gg 1$ (when the transient processes are finished) we have from equation (40)

$$\mathcal{N}_m(t) \approx \left(\frac{a\nu}{2a+\nu}\right)^m \mathrm{e}^{-\lambda_m t} \qquad \mathcal{Q}(t) \approx -\frac{a\nu}{2a+\nu}\mathrm{e}^{-D_1 t}[1 - \mathrm{e}^{-2D_2 t}].$$

For $\nu \gg a \gg 1$ we recover equations (43) and (59). In the opposite case $a \gg \nu + n - 1$, the initial value of the mean photon number disappears from the coefficients A_n, and we arrive at (22).

5.2. Binomial and Fock's states

The *binomial state* [18–22] has the following photon distribution and the generating functions (M is an integer, $0 \leqslant \eta \leqslant 1$):

$$p_n^{(\mathrm{bin})} = \binom{M}{n} \eta^n (1-\eta)^{M-n} \qquad F_0^{(\mathrm{bin})}(z) = (1-\eta+\eta z)^M.$$

Its factorial moments and Mandel's parameter read

$$\bar{n}_0 = M\eta \qquad \mathcal{N}_m = \frac{M!}{(M-m)!}\eta^m \qquad \mathcal{Q} = -\eta. \tag{62}$$

In this case, Mandel's parameter does not depend on the mean photon number, and it is always negative. The coefficients A_n can be expressed in terms of the Jacobi polynomials or the Gauss hypergeometric function $F(a, b; c; z)$ (see the appendix):

$$\begin{aligned} A_n^{(\mathrm{bin})}(M,\eta) &= \frac{M!(2\eta)^M}{(n-1+\nu)_n(2n+\nu)_{M-n}} P_{M-n}^{(-M,-M-\nu)}\left(\frac{\eta-1}{\eta}\right) \\ &= (2\eta)^n \frac{(M-n+1)_n}{(M+\nu)_n} \frac{2n-1+\nu}{n-1+\nu} F(n-M, n; 1-M-\nu; 1-2\eta). \end{aligned} \tag{63}$$

If both M and ν are large, then it is not difficult to obtain the asymptotics of (63),

$$A_n^{(\mathrm{bin})}(M,\eta) \approx \left(\frac{2M\eta}{2M\eta+\nu}\right)^n$$

which coincides with (61), if one takes into account that $M\eta = \bar{n}_0$.

If $\eta = 1$, we have the initial *Fock state* $|M\rangle$: $p_n(0) = \delta_{nM}$, $F_0(z) = z^M$. For $M \geqslant n \geqslant 1$,

$$\begin{aligned} A_n^{(\mathrm{Fock})}(M) &= \frac{M!2^M}{(n-1+\nu)_n(2n+\nu)_{M-n}} P_{M-n}^{(-M,-M-\nu)}(0) \\ &= 2^n \frac{(M-n+1)_n}{(M+\nu)_n} \frac{2n-1+\nu}{n-1+\nu} F(n-M, n; 1-M-\nu; -1). \end{aligned} \tag{64}$$

However, the simplest expressions can be obtained in the case $\eta = \frac{1}{2}$, when the hypergeometric function equals unity:

$$A_n^{(\mathrm{bin})}(M, \tfrac{1}{2}) = \frac{(M-n+1)_n}{(M+\nu)_n} \frac{2n-1+\nu}{n-1+\nu}. \tag{65}$$

Explicit expressions in terms of the factorials and elementary functions can be written also for integral values of the parameter ν. For example,

$$A_n^{(\mathrm{bin})}(M, \nu=1, \eta=\tfrac{1}{2}) = \frac{2(M!)^2}{(M-n)!(M+n)!}.$$

5.3. Negative binomial and thermal states

The *negative binomial states* are described by means of the relations ($\mu > 0$, $0 \leqslant \xi < 1$)

$$p_n(\mu,\xi) = (1-\xi)^\mu \frac{(\mu)_n}{n!}\xi^n \qquad F_0(z) = \left(\frac{1-\xi}{1-z\xi}\right)^\mu$$

$$\bar{n}_0 = \frac{\mu\xi}{1-\xi} \qquad \mathcal{N}_m = (\mu)_m \left(\frac{\xi}{1-\xi}\right)^m \qquad \mathcal{Q} = \frac{\xi}{1-\xi}.$$

These states were discussed in [20, 23]. The special case $\mu = 1$ corresponds to the initial *thermal state* with the dimensionless inverse temperature $1/T = \ln(1/\xi)$:

$$p_n(\xi) = (1-\xi)\xi^n \qquad \bar{n} = \mathcal{Q} = \frac{\xi}{1-\xi} \qquad F_0(z) = \frac{1-\xi}{1-z\xi}.$$

For these states we have the following coefficients (see the appendix):

$$A_n^{(\text{negbin})} = \frac{(\mu)_n}{(n-1+\nu)_n}\left(\frac{2\xi}{1-\xi}\right)^n F\left(\mu+n, n; 2n+\nu; \frac{-2\xi}{1-\xi}\right)$$
$$= \frac{(\mu)_n(2n-1+\nu)}{(n-1)!(n-1+\nu)}(2\bar{n}_0/\mu)^n \int_0^1 t^{n-1}(1-t)^{n-1+\nu}(1+2t\bar{n}_0/\mu)^{-n-\mu}\,\mathrm{d}t. \tag{66}$$

In the special case $\nu = 0$, $\mu = 1$, the Gauss hypergeometric function admits a quadratic transformation, and (66) can be written in the same form as in [24], where the evolution of the thermal states under pure two-photon processes was considered. When $\xi \to 1$ ($\bar{n}_0 \to \infty$), ξ disappears from the right-hand side of equation (66) (see asymptotics of the hypergeometric function in the appendix), and we arrive again at the universal formula (22).

For some specific values of parameters, the explicit expressions for A_n in terms of the elementary functions exist. For example,

$$A_n^{(\text{negbin})}(\mu = \tfrac{1}{2}, \nu = 1) = 2(2\xi)^n\left[\sqrt{1-\xi}+\sqrt{1+\xi}\right]^{-2n}.$$

Figure 4 demonstrates the time dependence of the mean photon number for the coherent states and for the special cases of binomial and negative binomial states with $\eta = \mu = \frac{1}{2}$.

If parameters μ and ν and integers, then the hypergeometric function of argument $-y$ can be expressed in terms of a finite number of derivatives of function $y^{-1}\ln(1+y)$ (see the appendix). However, in numerical calculations it is better to use the integral representation of coefficients A_n given in the second line of formula (66).

5.4. *Even and odd coherent states*

Now we proceed to the superposition states possessing extremal initial values of the parity coefficients: $b_\pm(0) = 0$ or 1. Let us begin with *even and odd coherent states* introduced in [25] (later publications can be found in [26, 27]; for the most recent generalizations see, e.g. [28–30]):

$$|\alpha; \pm\rangle = [2(1 \pm \mathrm{e}^{-2a})]^{-1/2}(|\alpha\rangle \pm |-\alpha\rangle) \qquad a = |\alpha|^2$$
$$p_{2n}^{(+)} = \frac{a^{2n}}{(2n)!\cosh a} \qquad p_{2n+1}^{(+)} = 0 \qquad F_0^{(+)}(z) = \frac{\cosh(az)}{\cosh a}$$
$$p_{2n+1}^{(-)} = \frac{a^{2n+1}}{(2n+1)!\sinh a} \qquad p_{2n}^{(-)} = 0 \qquad F_0^{(-)}(z) = \frac{\sinh(az)}{\sinh a}.$$

The initial factorial moments read

$$\mathcal{N}_{2k+1}^{(+)} = a^{2k+1}\tanh a \qquad \mathcal{N}_{2k+1}^{(-)} = a^{2k+1}\coth a \qquad \mathcal{N}_{2k}^{(\pm)} = a^{2k}.$$

Mandel's parameter assumes opposite signs for the even and odd states, and its absolute value does not exceed unity:

$$\mathcal{Q}^{(\pm)} = \pm\frac{2a}{\sinh(2a)}.$$

Using equation (57) we obtain the following coefficients of the expansion (9):

$$A_n^{(\pm)} = \frac{\Gamma(n-1+\nu)}{\Gamma(2n-1+\nu)} \frac{(2a)^n}{e^a \pm e^{-a}} [e^a \Phi(n; \nu+2n; -2a) \pm (-1)^n e^{-a} \Phi(n; \nu+2n; 2a)].$$

If $1 \sim a \ll \nu$, the mean photon number is given by equation (58), both for even and odd states. However, the relations between the parameters a and $\bar{n}_0$ are different. Therefore, having the same initial value $\bar{n}_0$, at $t > 0$ we obtain $\bar{n}^{(+)}(t) < \bar{n}^{(\text{coh})}(t) < \bar{n}^{(-)}(t)$. For Mandel's parameter we obtain the expressions (compare with equation (59))

$$\mathcal{Q}^{(+)}(t) = \frac{2a}{\sinh(2a)} e^{-D_1 t} [e^{-2D_2 t} \cosh^2(a) - \sinh^2(a)]$$

$$\mathcal{Q}^{(-)}(t) = -\frac{2a}{\sinh(2a)} e^{-D_1 t} [\cosh^2(a) - \sinh^2(a) e^{-2D_2 t}]$$

the corrections are of the order of a/ν:

If $a \sim \nu \gg 1$, then $\Phi(n; \nu+2n; 2a)$ tends to zero [31]:

$$\Phi(n; \nu+2n; 2a) \sim \begin{cases} e^{-2a}[(\nu/2a) - 1]^{-n} & \text{for } \nu/2a > 1 \\ (2a/\nu^2)^n \exp[-2a + c\nu - \nu \ln \nu] & \text{for } a > \nu \end{cases}$$

($c \sim 1$ being some function of the ratio a/ν). In this case the expressions for the coefficients A_n in the initial even and odd states practically coincide with formula (57) for the coherent states. However, for moderate values of a, ν, and $D_{1,2}t$, a nontrivial behaviour of the $\mathcal{Q}$-factor is observed. Figure 5 shows the dependence $\mathcal{Q}(t)$ for coherent, even, and odd states with the same initial mean photon number $\bar{n}_0 = 4$. Although the initial values of $\mathcal{Q}$-factor are very close, $\mathcal{Q}^{(\text{coh})}(0) = 0$, $\mathcal{Q}^{(\pm)}(0) \approx \pm 0.005$, the evolution turns out quite different in the time interval $0.1 < t < 1$. Moreover, the plot of function $\mathcal{Q}^{(\text{clas})}(t)$ calculated on the basis of the interpolation (56) (not shown in the figure) differs significantly from any exact curve.

5.5. *Even and odd binomial states*

These states are described by the formulae

$$F_0^{(\pm)}(z) = \frac{(1-\eta+\eta z)^M \pm (1-\eta-\eta z)^M}{1 \pm (1-2\eta)^M}$$

$$p_n^{(\pm)} = \binom{M}{n} \frac{(1-\eta)^{M-n} \eta^n [1 \pm (-1)^n]}{1 \pm (1-2\eta)^M}$$

$$\mathcal{N}_m^{(\pm)} = \frac{M!}{(M-m)!} \eta^m \frac{1 \pm (-1)^m (1-2\eta)^{M-m}}{1 \pm (1-2\eta)^M}.$$

The even binomial states were introduced in [32]. For the coefficients A_n we obtain the expression

$$A_n^{(\pm)}(M, \eta) = \frac{(2\eta)^n}{1 \pm (1-2\eta)^M} \frac{(M-n+1)_n}{(M+\nu)_n} \frac{2n-1+\nu}{n-1+\nu} \left[F(n-M, n; 1-M-\nu; 1-2\eta) \right.$$
$$\left. \pm (-1)^n (1-2\eta)^{M-n} F\left(n-M, n; 1-M-\nu; \frac{1}{1-2\eta}\right) \right].$$

We confine ourselves to the case of $\eta = \frac{1}{2}$, when all the formulae simplify significantly:

$$p_n^{(\pm 1/2)} = 2^{-M} \binom{M}{n} [1 \pm (-1)^n]$$

$$A_n^{(\pm)}(M,\tfrac{1}{2}) = \frac{(M-n+1)_n}{(M+\nu)_n}\frac{2n-1+\nu}{n-1+\nu}\left[1\pm(-1)^n\frac{(M-1)!\Gamma(n+\nu)}{(n-1)!\Gamma(M+\nu)}\right].$$

The peculiarity of this case is that the initial $\mathcal{Q}$-factor and all but one factorial moments are given by the same expressions (62) as for the original binomial states, excepting the Mth factorial moment:

$$\mathcal{N}_M^{(\pm)}(\eta=\tfrac{1}{2}) = M!2^{-M}[1\pm(-1)^M].$$

None the less, since the two-photon processes are sensitive to the high-order factorial moments, the $\mathcal{Q}$-factor begins to feel the difference in this last moment rather quickly: see figure 5. It is interesting that after a short transient time, the curves corresponding to the same initial parity of states (even, odd, or states without definite parity) become very close (and well separated from the curves with different initial parities), although the initial values $\mathcal{Q}(0)$ are quite different.

5.6. *Even and odd negative binomial states*

Even and odd counterparts of negative binomial states are described by the relations

$$F^{(\pm)}(z;\mu,\xi) = \mathcal{B}^{(\pm)}(\mu,\xi)[(1-z\xi)^{-\mu}\pm(1+z\xi)^{-\mu}]$$

$$p_n^{(\pm)}(\mu,\xi) = \mathcal{B}^{(\pm)}(\mu,\xi)\frac{(\mu)_n}{n!}\xi^n[1\pm(-1)^n]$$

$$\mathcal{N}_m^{(\pm)}(\mu,\xi) = \mathcal{B}^{(\pm)}(\mu,\xi)\mu_m\xi^m[(1-\xi)^{-\mu-m}\pm(-1)^m(1+\xi)^{-\mu-m}]$$

with the normalization constant

$$\mathcal{B}^{(\pm)}(\mu,\xi) = [(1-\xi)^{-\mu}\pm(1+\xi)^{-\mu}]^{-1}.$$

For these states the coefficients $A_n^{(\pm)}$ read

$$A_n^{(\pm)} = \frac{(\mu)_n}{(n-1+\nu)_n(2\xi)^n}\mathcal{B}^{(\pm)}(\mu,\xi)\left[(1-\xi)^{-\mu-n}F\left(\mu+n,n;2n+\nu;\frac{2\xi}{\xi-1}\right)\right.$$
$$\left.\pm(-1)^n(1+\xi)^{-\mu-n}F\left(\mu+n,n;2n+\nu;\frac{2\xi}{\xi+1}\right)\right].$$

In the special case of *even and odd thermal states* (the even thermal states were considered in [33, 34]) we have ($\mu = 1$)

$$p_n^{(\pm)} = \tfrac{1}{2}g_\pm(1-\xi^2)\xi^n[1\pm(-1)^n] \qquad g_+ = 1 \qquad g_- = 1/\xi$$

$$F_{\text{therm}}^{(+)}(z) = \frac{1-\xi^2}{1-z^2\xi^2} \qquad F_{\text{therm}}^{(-)}(z) = z\frac{1-\xi^2}{1-z^2\xi^2}$$

$$\overline{n}^{(+)} = \frac{2\xi^2}{1-\xi^2} \qquad \xi^2 = \frac{\overline{n}^{(+)}}{\overline{n}^{(+)}+2} \qquad \overline{n}^{(-)} = \frac{1+\xi^2}{1-\xi^2} \qquad \xi^2 = \frac{\overline{n}^{(-)}-1}{\overline{n}^{(-)}+1}$$

$$\mathcal{Q}^{(+)} = \frac{1+\xi^2}{1-\xi^2} = \overline{n}^{(+)}+1 \qquad \mathcal{Q}^{(-)} = \frac{\xi^4+4\xi^2-1}{1-\xi^4} = \overline{n}^{(-)}-1-1/\overline{n}^{(-)}$$

$$\mathcal{N}_m^{(\pm)} = \frac{1}{2}m!g_\pm\left(\frac{\xi}{1-\xi^2}\right)^m[(1+\xi)^{m+1}\pm(-1)^m(1-\xi)^{m+1}]$$

$$A_n^{(\pm)} = \frac{n!(2\xi)^n(1-\xi^2)g_\pm}{2(n-1+\nu)_n}\left[(1-\xi)^{-n-1}F\left(1+n,n;2n+\nu;\frac{2\xi}{\xi-1}\right)\right.$$
$$\left.\pm(-1)^n(1+\xi)^{-n-1}F\left(1+n,n;2n+\nu;\frac{2\xi}{\xi+1}\right)\right].$$

In the limiting case $\mu \to 0$ of the odd negative binomial states go to the *odd logarithmic states*, which can be considered as an odd counterpart of the 'logarithmic states' introduced in [35]. These states have the following properties:

$$F(z;\xi) = \mathcal{G}(\xi)\ln\frac{1+z\xi}{1-z\xi} \qquad \mathcal{G}(\xi) = \left(\ln\frac{1+\xi}{1-\xi}\right)^{-1}$$

$$p_n = \mathcal{G}(\xi)\frac{\xi^n}{n}[1-(-1)^n] \qquad n \geqslant 1$$

$$\mathcal{N}_m = \mathcal{G}(\xi)(m-1)!\xi^m[(1-\xi)^{-m} - (-1)^m(1+\xi)^{-m}]$$

$$\overline{n} = \frac{2\xi\mathcal{G}(\xi)}{1-\xi^2} \qquad \overline{n(n-1)} = \frac{4\xi^3\mathcal{G}(\xi)}{(1-\xi^2)^2} \qquad \mathcal{Q} = \frac{2\xi}{1-\xi^2}[\xi - \mathcal{G}(\xi)]$$

$$A_n = \frac{(n-1)!(2\xi)^{-n-1}}{(n-1+\nu)_n}\left[(1-\xi)^{-n}F\left(n,n;2n+\nu;\frac{2\xi}{\xi-1}\right) - (-1)^n(1+\xi)^{-n}F\left(n,n;2n+\nu;\frac{2\xi}{\xi+1}\right)\right].$$

However, the plots of various functions (such as $\mathcal{Q}^{(\pm)}(t)$) are not so expressive as in the previous cases, so we do not bring them here. (For example, $\mathcal{Q}$-factor, being positive and large at $t = 0$, quickly goes to the negative domain and then slowly approaches zero value as $t \to \infty$.)

6. Summary and conclusions

Let us collate the main results of the paper. We have presented analytical solutions to the infinite set of coupled equations for the density matrix diagonal elements and for the factorial moments of all orders. The explicit expressions for the coefficients in these solutions are obtained for almost all interesting specific quantum states. The only exception is the squeezed state. For highly excited initial states, we have found simple interpolation formulae giving the time dependence of the factorial moments in the whole interval $0 < t < \infty$. These formulae are in a good agreement with the exact results even for rather moderate values of the initial factorial moments. We have demonstrated that Mandel's $\mathcal{Q}$-parameter is sensitive even to small differences in the initial values of the factorial moments, and it exhibits a nontrivial behaviour at the intermediate stage of the relaxation process, corresponding to the maximal competition between the one- and two-photon absorption. We have analysed in detail the universal distributions which arise for any highly excited initial state after a short transition time of the order of $(D_2\bar{n}_0)^{-1}$. Also, we have considered the influence of the ratio D_1/D_2 on the mean transition time from the initial state to the ground state, and we have found that this time remains finite even for highly excited states, provided that the two-photon absorption coefficient D_2 does not equal zero.

It should be noted that all the characteristics of the photon distributions considered above depend on the diagonal elements of the density matrix only. Therefore they coincide, e.g. for the genuine *pure* states and for the statistical mixtures with the same values of the diagonal matrix elements. For these reasons, it is interesting to study the behaviour of the *off-diagonal* matrix elements, which observes the difference between pure and mixed states (for pure two-photon absorption this was done in [36, 37]). We shall consider this problem in a separate paper.

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Appendix

Here we have collected the formulae used in deriving the relations given in sections 2–5. The division into sections is made in accordance with the main part of the paper.

A.1. From section 2

Equation (12) can be derived from formula 7.391.10 of [14],

$$\int_{-1}^{1}(1-x)^{\rho}(1+x)^{\beta}P_n^{(\alpha,\beta)}(x)P_m^{(\rho,\beta)}(x)\,\mathrm{d}x = \frac{2^{\rho+\beta+1}\Gamma(\rho+m+1)\Gamma(\beta+n+1)\Gamma(\alpha+\beta+m+n+1)}{m!(n-m)!\Gamma(\alpha+\beta+n+1)\Gamma(\rho+\beta+m+n+2)} \times\frac{\Gamma(\alpha-\rho-m+n)}{\Gamma(\alpha-\rho)}$$

(which holds for $\beta > -1$, $\rho > -1$), if one puts $\alpha = -1$, $\rho = -1+\varepsilon$, and takes the limit $\varepsilon \to 0$.

Rodrigues' formula for the Jacobi polynomials reads

$$(-1)^n 2^n n!(1-x)^{\alpha}(1+x)^{\beta}P_n^{(\alpha,\beta)}(x) = \frac{\mathrm{d}^n}{\mathrm{d}x^n}[(1-x)^{\alpha+n}(1+x)^{\beta+n}].$$

The Jacobi polynomial $P_k^{(\alpha,\beta)}(x)$ is expressed in terms of the Gauss hypergeometric function as follows [15]:

$$P_k^{(\alpha,\beta)}(x) = \frac{\Gamma(\beta+\alpha+2k+1)}{k!\Gamma(\beta+\alpha+k+1)}\left(\frac{x-1}{2}\right)^k F\left(-k,-k-\alpha;-2k-\alpha-\beta;\frac{2}{1-x}\right) = \binom{k+\beta}{k}\left(\frac{x-1}{2}\right)^k F\left(-k,-k-\alpha;\beta+1;\frac{x+1}{x-1}\right).$$

The partial cases of the hypergeometric function [38]:

$$F(a,b;a-b+1;-1) = \frac{2^{-a}\Gamma(\frac{1}{2})\Gamma(1+a-b)}{\Gamma(1+a/2-b)\Gamma(\frac{1}{2}+a/2)} \tag{A.1}$$

$$F(a,b;a-b+2;-1) = \frac{\Gamma(\frac{1}{2})\Gamma(2+a-b)}{2^a(b-1)} \times\left[\frac{1}{\Gamma(a/2)\Gamma(\frac{3}{2}+a/2-b)} - \frac{1}{\Gamma(\frac{1}{2}+a/2)\Gamma(1+a/2-b)}\right]. \tag{A.2}$$

The following known relations are useful in calculations:

$$(a)_n \equiv \frac{\Gamma(a+n)}{\Gamma(a)} \equiv a(a+1)\ldots(a+n-1) \qquad (a)_0 \equiv 1$$

$$(\xi-n)_k = (-1)^k(n-\xi-k+1)_k = (-1)^k\frac{\Gamma(n-\xi+1)}{\Gamma(n-\xi-k+1)}$$

$$\Gamma(2z) = 2^{2z-1}\Gamma(z)\Gamma(z+\tfrac{1}{2})/\Gamma(\tfrac{1}{2}).$$

A.2. From section 4

The Euler–Maclaurin summation formula up to the third-order terms reads [38]

$$\sum_{k=1}^{n-1} f_k = \int_0^n f(k)\,\mathrm{d}k - \tfrac{1}{2}[f(0) + f(n)] - \tfrac{1}{12}[f'(0) - f'(n)] + \tfrac{1}{720}[f'''(0) - f'''(n)] + \cdots. \tag{A.3}$$

In our case $n = \infty$, but function $f(n)$ and all its derivatives turn into zero when $n \to \infty$.

A.3. From section 5.1

Equation (57) is a consequence of the expansion ($-1 < x < 1$) [39]

$$\mathrm{e}^{xy} = (2y)^{-1-(\alpha+\beta)/2} \sum_{n=0}^{\infty} \frac{\Gamma(\alpha+\beta+n+1)}{\Gamma(\alpha+\beta+2n+1)} M_{\kappa,\mu}(2y) P_n^{(\alpha,\beta)}(x)$$

where $\kappa = (\alpha - \beta)/2$, $\mu = n + (\alpha + \beta + 1)/2$, and $M_{\kappa,\mu}(z)$ is the Whittaker function: $M_{\kappa,\mu}(z) = z^{\mu+1/2}\mathrm{e}^{-z/2}\Phi(\mu - \kappa + \frac{1}{2}; 1 + 2\mu; z) = z^{\mu+1/2}\mathrm{e}^{z/2}\Phi(\mu + \kappa + \frac{1}{2}; 1 + 2\mu; -z)$.

The confluent hypergeometric function $\Phi(a; c; x)$ has the following asymptotical expansions for large values of $|x|$ and fixed a and c [15]:

$$\Phi(a; c; x) \sim \begin{cases} \dfrac{\Gamma(c)}{\Gamma(a)} \mathrm{e}^x x^{a-c} & x > 0 \\ \dfrac{\Gamma(c)}{\Gamma(c-a)} (-x)^{-a} & x < 0. \end{cases}$$

Its integral representation reads

$$\Phi(a; c; x) = \frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)} \int_0^1 \mathrm{e}^{xu} u^{a-1} (1-u)^{c-a-1}\,\mathrm{d}u \qquad c > a > 0.$$

A.4. From section 5.2

The formulae in section 5.2 are based on the results of Carlson's monograph [40]. He gives the expansion (p 223, example 7.2-2)

$$(A + Bx)^M = \sum_{n=0}^{M} \binom{M}{n} B^n R_{M-n}(1 + \alpha + n, 1 + \beta + n; A - B, A + B) \times R_n(-\alpha - n, -\beta - n; x + 1, x - 1)$$

where his polynomials $R_n(f, f'; x, y)$ are related to the usual Jacobi polynomials as follows,

$$R_n(f, f'; x, y) = \frac{n!}{(f + f')_n} (y - x)^n P_n^{(-f-n,\,-f'-n)}\left(\frac{x+y}{x-y}\right).$$

Thus we obtain the formula

$$(A + Bx)^M = \sum_{n=0}^{M} \frac{M!(2B)^M}{(n + \alpha + \beta + 1)_n (2n + \alpha + \beta + 2)_{M-n}} \times P_{M-n}^{(-1-M-\alpha,-1-M-\beta)}\left(-\frac{A}{B}\right) P_n^{(\alpha,\beta)}(x).$$

Equation (63) is its special case for $\alpha = -1$, $\beta = -1 + \nu$, $B = 1 - A = \eta$.

A.5. From section 5.3

In the case of the negative binomial states we use formula ([40], p 227, example 7.7-14)

$$(A-Bx)^{-\mu}=\sum_{n=0}^{\infty}\frac{(\mu)_n}{n!}B^nR_{-\mu-n}(1+\alpha+n,1+\beta+n;A+B,A-B)$$
$$\times R_n(-\alpha-n,-\beta-n;x+1,x-1).$$

With the aid of the relation ([40], p 117)

$$R_{-a}(f,f';x,y)=y^{-a}F\left(a,f;f+f';1-\frac{x}{y}\right)$$

we can write

$$(A-Bx)^{-\mu}=\sum_{n=0}^{\infty}\frac{(\mu)_n(2B)^nF(\mu+n,1+\alpha+n;2+\alpha+\beta+2n;\frac{2B}{B-A})}{(A-B)^{\mu+n}(1+\alpha+\beta+n)_n}P_n^{(\alpha,\beta)}(x).$$

Taking $\alpha=-1$, $\beta=-1+\nu$, $A=1$, $B=\xi$, we arrive at equation (66).

The integral representation for the Gauss hypergeometric function ($c>b>0$):

$$F(a,b;c;z)=\frac{\Gamma(c)}{\Gamma(b)\Gamma(c-b)}\int_0^1 t^{b-1}(1-t)^{c-b-1}(1-tz)^{-a}\,\mathrm{d}t. \qquad \text{(A.4)}$$

The leading term of the asymptotical expansion of $F(\mu+n,n;2n+\nu;-z)$ for $z\gg 1$ and $\mu>0$ reads [15]

$$F(\mu+n,n;2n+\nu;-z)\approx\frac{\Gamma(2n+\nu)}{(\mu)_n\Gamma(n+\nu)}z^{-n}.$$

If μ and ν are nonnegative integers, then formula 2.2.2(4) from [15] yields

$$F(n+\mu,n;2n+\nu;-y)=\frac{(-1)^{\mu+\nu}(2n+\nu-1)!}{(n+\nu-1-\mu)!(n-1)!(n+\mu-1)!(n+\nu-1)!}$$
$$\times\hat{D}^{n-1+\mu}\left((1+y)^{n-1+\nu}\hat{D}^{n-1+\nu-\mu}\left[\frac{1}{y}\ln(1+y)\right]\right)$$

where $\hat{D}\equiv \mathrm{d}/\mathrm{d}y$.

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