Squeezing of a coherent pump field during hyper-Raman scattering

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The possibility of a conversion of a coherent pump field into squeezed light in the course of hyper-Raman scattering is discussed. As a result of the nonlinear interaction with the medium, the pump field undergoes a pronounced squeezing. The extent of this squeezing is considerably greater than that of the corresponding squeezing in other nonlinear processes involving a two-photon absorption of a pump. Results are derived in a quantum theory of hyper-Raman scattering through an exact solution of the equation for the density matrix of two modes: the pump and the Stokes radiation.

1. INTRODUCTION

Squeezed light has been produced successfully in a number of nonlinear optical processes (Refs. 1–4; see also Ref. 5). However, the extent of squeezing which can be observed is only slight, and many aspects of nonclassical light are exhibited in a weak field, disappearing in the case of intense generation. These circumstances make it necessary to seek new ways to generate intense, greatly squeezed light. Of particular interest in this connection are hyper-Raman scattering (HRS) and the four-wave mixing (FWM) which it induces (Fig. 1).

We recall that Stokes radiation is generated in HRS in a two-photon–absorbing medium. This Stokes radiation acts in turn along with the pump to generate, by FWM, a third wave at the frequency of the transition $3 \to 1$. One reason why these processes have attracted interest is that the parametric coupling of the Stokes radiation with the third wave should lead to a squeezing of quantum fluctuations in both modes (or in the composite mode which they form). A second reason for the interest is that the pump field itself is squeezed, because of a nonlinear (two-photon) interaction with the medium. With different squeezing mechanisms, it becomes possible to study either effect in a setting in which it is completely independent of the other effect, with the pump field being interpreted at the classical level in the former case and at the quantum-mechanical level in the latter.

In this paper we are reporting a study of squeezing effects in a coherent pump field; corresponding calculations for the Stokes and parametric waves were carried out in Ref. 6. From the standpoint of the problem posed above, the results of the theory which we have derived are fairly convincing: The extent of the squeezing of the pump field in the course of HRS can be more than 80% of the maximum value permitted by quantum mechanics. We recall that we are talking not about a *generation* of squeezed light, in which case the light would usually be of low intensity, but about a conversion of an intense coherent pump field into squeezed light.

Field squeezing in the course of a nonlinear interaction of the field with a medium was discussed in Refs. 7–10 in the cases of harmonic generation[7,8] and two-photon absorption of coherent light.[9,10] A comparison of the results of those previous studies with our own results shows that among the various processes which involve a two-photon absorption of a pump the greatest squeezing of a laser-light field would be expected in the case of HRS.

2. BASIC EQUATIONS OF THE SYSTEM

We consider the propagation of several pulses through a medium of four-level atoms (Fig. 1): a pump pulse at a frequency ω_1; Stokes radiation, which is generated at the frequency ω_2 in the course of hyper-Raman scattering; and a parametric wave, generated at a frequency $\omega_3 = 2\omega_1 - \omega_2 = \omega_{31}$ through four-wave mixing. Since we are dealing with a forward propagation of pulses, for which the correspondence $t \leftrightarrow z/c$ holds, we will analyze the evolution of the pulses as a function of t. The fields of all three waves are expressed in terms of annihilation (creation) operators $a(a^+)$, $b(b^+)$, and $c(c^+)$ as follows:

$$E_1^{(+)} = -i\left(\frac{2\pi\hbar\omega_1}{V}\right)^{1/2} a \exp(ik_1 z - i\omega_1 t),$$

$$E_{2(3)}^{(+)} = E_1^{(+)} \, [1 \to 2(3), \quad a \to b(c)], \tag{1}$$

where V is the quantization volume.

The effective interaction Hamiltonian is then

$$H = \hbar \sum_i [G a^2 b^+ \exp(i\Delta k z_i) + fc] \exp(ik_3 z_i) \sigma_{31}^{(i)} + \text{H.a.},$$

$$G = -\frac{i}{\hbar}\left(\frac{2\pi\hbar}{V}\right)^{3/2} \omega_1 \omega_2^{1/2} \mu_{23} \sum_n \frac{\mu_{1n}\mu_{n2}}{\hbar^2 \Delta_{n1} \Delta},$$

$$f = -\frac{i}{\hbar}\left(\frac{2\pi\omega_3\hbar}{V}\right)^{1/2} \mu_{31}, \tag{2}$$

where μ_{ij} is the dipole matrix element of the atomic transition $i \to j$, $\Delta k = 2k_1 - k_2 - k_3$, $\sigma_{31}^{(i)}$ is an element of the den-

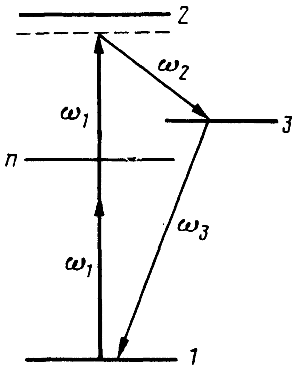

FIG. 1. Scheme of the interaction in hyper-Raman scattering.

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sity matrix of atom i corresponding to the $1\rightarrow 3$ transition current, $\Delta=\omega_{21}-2\omega_1$ is the two-photon detuning, and $\Delta_{n1}=\omega_{n1}-\omega_1$. We are assuming that all the intermediate states n over which the summation is carried out in the expression for G are far from a one-photon resonance with the pump field.

We describe the time evolution of the fields by means of an equation for the density matrix ρ of three modes. This equation is constructed in the standard way,[11] through the use of Hamiltonian (2) and under the assumption that the atoms are always in ground state 1:

$$\partial\rho/\partial t=-\mathcal{K}[a^{+2}a^2bb^+\rho-2a^2b^+\rho a^{+2}b+\rho a^{+2}a^2bb^+]$$
$$-\frac{\alpha_c}{2}[c^+c\rho-2c\rho c^+$$
$$+\rho c^+c]+\{\beta[a^2b^+c^+\rho-2a^2b^+\rho c^++\rho a^2b^+c^+]+\text{H.a.}\}. \quad (3)$$

The first term in (3) describes the HRS, i.e., the absorption of two pump photons and the emission of one photon in the Stokes mode. The quantity

$$\mathcal{K}=g(2\omega_1-\omega_2)|G|^2\int_V N(\mathbf{r})d^3r$$

is related to the gain for the Stokes radiation. This relationship follows directly from the equation for the average number $\langle n_b\rangle$, of photons in mode b, which is found with the help of Eq. (3), in which the amplitude of the pump field is assumed to be a c-number and given. We can then write

$$\partial\langle n_b\rangle/\partial t=\alpha_b(\langle n_b\rangle+1)+\ldots,$$

where $\alpha_b=2\mathcal{K}n_a$ is the gain for the Stokes radiation, and n_a is the initial number of photons in the pump mode.

The second term in (3) corresponds to the absorption of a c-mode photon with an absorption coefficient

$$\alpha_c=2g(\omega_3)|f|^2\int_V N(\mathbf{r})d^3r.$$

The last term corresponds to a parametric interaction between the fields, with a coupling constant

$$\beta=-g(\omega_3)Gf^*\int_V N(\mathbf{r})d^3r.$$

Here $g(\omega_3)$ is the shape of the atomic transition line at the frequency $\omega_3=\omega_{31}$. It can effectively be replaced by Γ^{-1}, the inverse width of the $3\rightarrow 1$ transition. Here $N(\mathbf{r})$ is the number density of atoms in the medium. In deriving (3) we used the phase-matching condition $\Delta k=0$.

We turn now to squeezing effects in the pump field.

3. TIME EVOLUTION OF THE SQUEEZING IN THE PUMP FIELD

Let us demonstrate how a squeezing of quantum fluctuations occurs in a coherent pump field during hyper-Raman scattering. The effect evidently becomes significant and observable in the case of stimulated HRS, in which the number of photons absorbed from the pump field and thus the number of Stokes photons are not small. According to Ref. 12, this situation corresponds to the case $\alpha_b\gg\alpha_c$, in which we can ignore the effect of c-mode generation on the HRS. Retaining on the right side of Eq. (3), only the first term, which corresponds to HRS, we then find an equation which describes the evolution of the density matrix for the pump mode and for the Stokes-radiation mode:

$$\partial\rho/\partial t=-\mathcal{K}[a^{+2}a^2bb^+\rho-2a^2b^+\rho a^{+2}b+\rho a^{+2}a^2bb^+]. \quad (4)$$

It is convenient to solve this equation for the matrix elements ρ_{mn}^{rk}, which are determined from the expression

$$\rho=\sum_{m,n,r,k}\rho_{mn}{}^{rk}|m,r\rangle\langle n,k|,$$

where the subscripts and superscripts in ρ_{mn}^{rk} refer to the a and b modes, respectively. For them, we find the following equation from (4):

$$\partial\rho_{mn}{}^{rk}/\partial t=-\mathcal{K}[(m^2-m)(r+1)+(n^2-n)(k+1)]\rho_{mn}{}^{rk}$$
$$+2\mathcal{K}[(m+1)(m+2)(n+1)(n+2)rk]^{1/2}\rho_{m+2,n+2}^{r-1,k-1}. \quad (5)$$

The initial state of the pump field is assumed to be a coherent state: $|z\rangle=||z|,\exp(i\theta)\rangle$. The Stokes field is assumed to be in the vacuum state. The boundary conditions on (5) are then

$$\rho_{mn}{}^{rk}(0)=\frac{1}{(m!n!)^{1/2}}(z^*)^m z^n\exp(-|z|^2)\delta_{r0}\delta_{k0}. \quad (6)$$

The expectation value of any pump-field operator $\hat{O}$ is calculated in accordance with

$$\langle\hat{O}\rangle=\mathrm{Sp}[\hat{O}\rho]=\sum O_{mn}\rho_{nm}{}^{rr},$$

so we will need only the matrix elements ρ_{mn}^{rr} below. Equation (5) can be solved for these elements exactly by means of Laplace transforms. This solution is found in the Appendix; it is given by a fairly complex expression. If we introduce the operator $\hat{Q}$, however, we can rewrite this solution in the compact form

$$\rho_{mn}{}^{rk}(T)=\frac{r!}{(m!n!)^{1/2}}|z|^{4r}(z^*)^m z^n$$
$$\times\exp(-|z|^2)\hat{Q}\sum_{q=0}^{r}\frac{\exp[-L_{mn}{}^r(q)T]}{\prod_{k=0,\,k\neq q}^{r}[L_{mn}{}^r(k)-L_{mn}{}^r(q)]},$$
$$L_{mn}{}^r(q)=\frac{q+1}{2}[m+2(r-q)][m+2(r-q)-1]$$
$$+\frac{q+1}{2}[n+2(r-q)][n+2(r-q)-1],\qquad T=2\mathcal{K}t, \quad (7)$$

where the effect of $\hat{Q}$ reduces to the following: If, for $k\neq q$, k, $q=0,1,\ldots,r$, none of the factors $L(k)-L(q)$ in (7) vanishes, then $\hat{Q}=1$, where $L(q)\equiv L^r_{mn}(q)$. For $L(q_1)=L(q_2)$; $q_1\neq q_2$; $q_1,q_2\in[0,r]$, however, we have

$$\hat{Q}\sum_{q=0}^{r}\frac{\exp[-L(q)T]}{\prod_{k=0,\,k\neq q}^{r}[L(k)-L(q)]}=\left(-\frac{\partial}{\partial L(q_1)}\right)$$
$$\times\sum_{q=0,\,q\neq q_2}^{r}\frac{\exp[-L(q)T]}{\prod_{k=0,\,k\neq q,q_2}^{r}[L(k)-L(q)]}. \quad (8)$$

If the number of vanishing factors in the denominator in (7) is greater than one, i.e., if it is $l>1$, then $\hat{Q}$ acts l times as a formal differentiation operator, by analogy with (8).

We will calculate the following parameters for the pump field:

1) The average number of photons in a mode,

$$\langle n_a(t)\rangle=\langle a^+(t)a(t)\rangle=\sum_{n,r} n\rho_{nn}{}^{rr}(t). \quad (9)$$

2) The squeezing parameter

$$S_a(t)=\frac{1}{4}\sum_{n,r}[n(n-1)]^{1/2}(\rho^{rr}_{n,n-2}+\rho^{rr*}_{n,n-2})+\frac{1}{2}\langle n_a(t)\rangle - \frac{1}{4}\left|\sum_{n,r} n^{1/2}(\rho^{rr}_{n,n-1}+\rho^{rr*}_{n,n-1})\right|^2. \quad (10)$$

If this parameter is a negative, the meaning is that the field component $(a+a^+)$ is squeezed. The value $S_a=-\frac{1}{4}$ corresponds to the maximum squeezing.

3) The second-order correlation function (G-factor)

$$G^{(2)}=\frac{\langle n_a^2(t)\rangle-\langle n_a(t)\rangle}{\langle n_a(t)\rangle^2}, \quad \langle n_a^2(t)\rangle=\sum_{n,r} n^2\rho_{nn}{}^{rr}(t). \quad (11)$$

The only way to find exact solutions for these quantities is to go through a numerical calculation of the sums in (9)–(11). We present the results of such a calculation below. However, so that we can see how these parameters evolve in time, we will derive solutions for them in the limits of small and large T; these solutions exist in analytic form.

a) Solution for small T.

Using (7), we can verify that an expansion of ρ^{rr}_{mn} in T begins with T^r, i.e., that

$$\rho_{mn}{}^{rr}(T)=\mathrm{const}\cdot T^r+O(T^{r+1})+\ldots, \quad T\ll 1. \quad (12)$$

This point can be demonstrated more easily by directly integrating Eq. (5) at small T. As a result we find

$$\rho_{mn}{}^{rr}(T)=r[(m+1)(m+2)(n+1)(n+2)]^{1/2}\int_0^T dT'\rho^{r-1,r-1}_{m+2,n+2}(T'),$$

and we can assume that at $T\ll 1$ we have $\rho^{00}_{mn}(T)\approx\rho^{00}_{mn}(0)$.

Since $\Sigma_m\rho^{rr}_{mm}$ is the probability that the number of photons in the Stokes mode is r, according to (12) this probability also increases as T^r during the first few moments after the interaction is turned on.

We find the parameters $\langle n_a\rangle$, $G^{(2)}$, and S_a through an expansion in T in which terms up to T^2 are retained. We can carry out this procedure if we retain, in the summation over r in (9)–(11), only the first three terms, with $r=0$, 1, and 2. Here we can show that we have $\hat{Q}\equiv 1$ in (7). The same result can be found more simply by working from the formula

$$\langle\hat{O}\rangle=\mathrm{Sp}\left\{\hat{O}\left[\rho(0)+T\dot{\rho}(0)+\frac{T^2}{2}\ddot{\rho}(0)\right]\right\},$$

where $\ddot{\rho}(T)$ is found from Eq. (4), after the latter is differentiated with respect to T one more time. We thus have

$$\langle n_a(T)\rangle=|z|^2-2|z|^4T+2\left[2|z|^6+|z|^4-\frac{|z|^8}{2}\right]T^2,$$

$$S_a(T)=|z|^4\frac{T^2}{2}+\frac{|z|^2\cos 2\theta}{2}\times\left[-T+\left(-\frac{|z|^4}{2}+3|z|^2+\frac{1}{2}\right)T^2\right],$$

$$G^{(2)}(T)=1-2T+[-|z|^4+4|z|^2+2]T^2. \quad (13)$$

It is interesting to compare these results with the corresponding expressions derived for n, S_a, and $G^{(2)}$ in the case of two-photon absorption (TPA) of coherent light.[9] A comparison yields

$$\langle n_a(T)\rangle_{\mathrm{HRS}}=\langle n_a(T)\rangle_{\mathrm{TPA}}-|z|^8T^2,$$

$$S_{\mathrm{HRS}}(T)=S_{\mathrm{TPA}}(T)-\frac{|z|^4}{4}T^2\cos 2\theta. \quad (14)$$

One can also show that $G^{(2)}_{\mathrm{HRS}}=G^{(2)}_{\mathrm{TPA}}-|z|^4T^2$. It follows that early in the process the pump field is absorbed and is squeezed to a greater extent during HRS than during two-photon absorption. The exact solution shows (see the discussion below) that this tendency persists even later, leading in the case of HRS to a significantly greater squeezing of the pump field. The distinction between the two processes starts to become apparent at terms on the order of T^2. This circumstance can be explained in a relatively simple way by noting that a distinguishing feature of HRS is the additional emission of Stokes photons. Since the probability for the appearance of the first photon in the Stokes mode is proportional to T, however, it comes into play in terms of order T^2 and higher.

b) Steady-state solution at long T.

After a long time the system should evidently reach a steady state, since the pump will be completely depleted. The system at this point either will contain only one photon, or it will go into the vacuum state. The reason is that, according to (7), the only matrix elements ρ^{rr}_{mn} which do not vanish in the limit $T\to\infty$ are those for which the condition $L^r_{mn}(q)=0$ holds. It is not difficult to verify that this condition holds if $q=r$ and if m, $n=0$ or 1, i.e., if only ρ^{rr}_{00}, ρ^{rr}_{11}, and $\rho^{rr}_{01}=(\rho^{rr}_{10})^*$ are nonzero. For them we find the following results from (7) in the limit $T\to\infty$:

$$\rho_{00}{}^{rr}(\infty)=|z|^{4r}\exp(-|z|^2)/(2r)!, \quad (15)$$

$$\rho_{11}{}^{rr}(\infty)=|z|^{2(2r+1)}\frac{\exp(-|z|^2)}{(2r+1)!}, \quad (16)$$

$$\rho_{01}{}^{rr}(\infty)=z\left(\frac{|z|^2}{2}\right)^{2r}\frac{\exp(-|z|^2)}{(r!)^2}. \quad (17)$$

Obviously,

$$\sum_r[\rho_{00}{}^{rr}(\infty)+\rho_{11}{}^{rr}(\infty)]=1.$$

Since Eq. (5) actually splits into two equations, one of which couples all the even-n ρ^{rr}_{nn} with each other, while the second does the same for the odd-n elements, and since we furthermore have $\Sigma_{n,r}\rho^{rr}_{nn}=1$, the following sums are conserved separately:

$$\sum_{n}^{\text{even}}\sum_{r}\rho_{nn}^{rr} \text{ and } \sum_{n}^{\text{odd}}\sum_{r}\rho_{nn}^{rr}.$$

Thus

$$\sum_{n}^{\text{even}}\sum_{r}\rho_{nn}^{rr}(\infty)=\sum_{n}^{\text{even}}\sum_{r}\rho_{nn}^{rr}(0),$$

$$\sum_{n}^{\text{odd}}\sum_{r}\rho_{nn}^{rr}(\infty)=\sum_{n}^{\text{odd}}\sum_{r}\rho_{nn}^{rr}(0). \quad (18)$$

Using (6) and (15), (16), we easily verify that these conservation laws hold for the solutions which we have found.

We now seek n_a, $G^{(2)}$, and S_a in steady state (15)–(17):

$$\langle n_a(\infty)\rangle=\sum_{r}\rho_{11}^{rr}(\infty)=\exp(-|z|^2)\times\text{sh}\,|z|^2. \quad (19)$$

Since the operator $a^+a/2+b^+b$ is conserved,

$$\frac{d}{dt}\left\langle\frac{a^+a}{2}+b^+b\right\rangle=0,$$

as is easily verified with the help of (4), we have

$$\frac{1}{2}\langle n_a(\infty)\rangle+\langle n_b(\infty)\rangle=\frac{1}{2}\langle n_a(0)\rangle+\langle n_b(0)\rangle.$$

Using (19) we find

$$\langle n_b(\infty)\rangle=\frac{1}{2}|z|^2-\frac{1}{4}[1-\exp(-2|z|^2)],$$

or, if $|z|^2\gg 1$,

$$\langle n_a(\infty)\rangle=\frac{1}{2} \text{ and } \langle n_b(\infty)\rangle=\frac{|z|^2}{2}.$$

For the squeezing parameter S_a we find

$$S_a(\infty)=\frac{1}{2}\exp(-|z|^2)\cdot\text{sh}|z|^2-\cos^2\theta\cdot|z|^2 \times\exp(-2|z|^2)\cdot I_0^2(|z|^2), \quad (20)$$

where

$$I_0(x)=\sum_{r}\left(\frac{x}{2}\right)^{2r}\frac{1}{(r!)^2}$$

is the modified Bessel function of index zero. For $|z|^2\gg 1$ we have

$$I_0(|z|^2)=\exp(|z|^2)/|z|(2\pi)^{1/2},$$

$$S_a(\infty)=\frac{1}{4}[1-\exp(-2|z|^2)]-\frac{\cos^2\theta}{2\pi},$$

i.e., $S_a(\infty)>0$. In the same limit we have $G^2(\infty)=0$, since

$$\langle n_a^2(\infty)\rangle=\sum_{n,r}n^2\rho_{nn}^{rr}(\infty)=\sum_{r}\rho_{11}^{rr}(\infty)=\langle n_a(\infty)\rangle.$$

This result was to be expected since in the limit $T\to\infty$ there is at best only one photon in the pump mode.

We turn now to the exact solutions for $\langle n_a\rangle$, $G^{(2)}$, and S_a. Figures 2–4 show the results of numerical calculations from Eqs. (9)–(11) and (7) for $\theta=0$. Shown for comparison are plots of $\langle n_a\rangle$, $G^{(2)}$ (Ref. 13), and S_a (Ref. 9) versus T in the case of two-photon absorption. We see that in the case of HRS the quantities $\langle n_a\rangle$ and $G^{(2)}$ fall off more rapidly with increasing T. These results are evidence of a more intense absorption (Fig. 2) and of a higher antibunching correlation (Fig. 3) in the pump mode than in the case of two-photon absorption. The distinction between the two processes can be seen far more vividly in the plot of $S_a(T)$ (Fig. 4). In the case of HRS, the minimum in S_a is much deeper and is shifted further down the T scale. This effect becomes greater with increasing initial number of photons in the coherent pump mode, $\bar{n}_a$. At large values of $\bar{n}_a(\sim 10)$ the extent of squeezing is more than 80% of the maximum possible extent, and it also exceeds the value of S_a predicted[7] in the case of second-harmonic generation. The following arguments reveal the reason for these differences among the three processes, each of which involves a two-photon absorption of a pump. This absorption must be most intense in the case of HRS, since its probability in this case, proportional to the number of Stokes photons, increases exponentially with T, while in the case of two-photon absorption it remains constant, and in the case of second-harmonic generation it increases only linearly with T, being proportional to the number of photons in the second-harmonic mode. It follows in particular from this assertion that the difference between two-photon absorption and HRS increases with increasing $\bar{n}_a$, since there is also an increase in the number of Stokes photons, as can indeed be seen in Fig. 4.

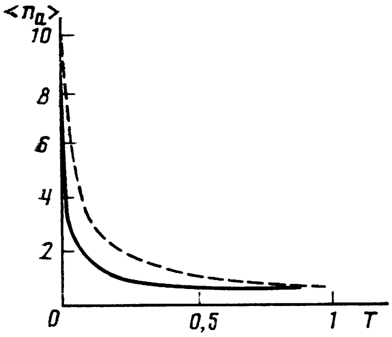

FIG. 2. Time evolution of the average number $\langle n_a\rangle$, of photons in the pump mode for an initial value $\bar{n}_a=10$. The dashed line shows the same function in the case of two-photon absorption, again for $\bar{n}_a=10$ (Ref. 13).

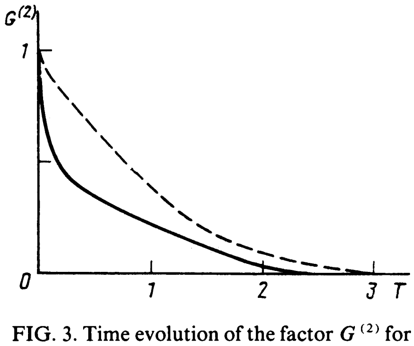

FIG. 3. Time evolution of the factor $G^{(2)}$ for the pump mode in the cases of (solid line) hyper-Raman scattering and (dashed line) two-photon absorption.[13]

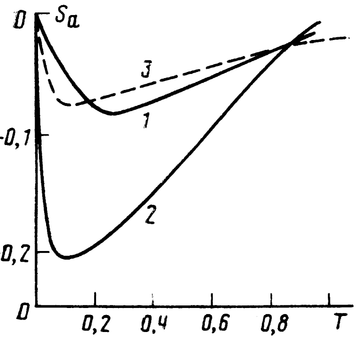

FIG. 4. The squeezing parameter S_a of the pump field versus T for the two values (1) $|z| = 1.73$ and (2) $|z| = 3$. Dashed line 3 represents the case of two-photon absorption[13] with $|z| = 3$.

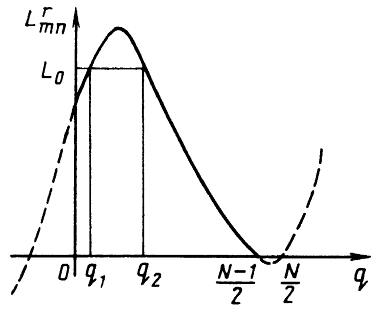

FIG. 5. L^r_{nn} versus q at a fixed value $N = n + 2r$. A given value L_0 is taken on at only two points, q_1 and q_2, which lie within the segment $[0, r_{\max}]$.

We have one final comment regarding the approximation, used above, that it is legitimate to ignore the effect of c-mode generation on the basis of FWM. Clearly, that approximation is not valid in the region of strong pump depletion or at large values of T. Incorporating that process does not alter the basic results (in particular, the prediction of a pronounced squeezing of the pump field), but it does lead to certain changes in the behavior of $\langle n_a \rangle$ and S_a as functions of T at large T. Accordingly, we will simply list these changes, omitting the detailed calculations. These changes occur because as the pump is depleted and, correspondingly, as the number of photons in the c-mode increases, the destructive interference between the two mechanisms for the filling of level 3 becomes complete, so that, no further absorption of the pump occurs. As a result, the new asymptotic values of $\langle n_a \rangle$, $G^{(2)}$, and S_a turn out to be larger than the previous values, in (19)–(20). The parameter S_a goes positive even at T values lower than those in Fig. 4; i.e., its minimum becomes narrower. All these effects, however, set in at a progressively later time as the ratio f_{32}/f_{13} inceases, where f_{ij} is the oscillator strength of the $i \to j$ transition. This fact must be taken into consideration in experimental tests of the theoretical predictions.

I wish to thank M. L. Ter-Mikaelyan and G. Yu. Kryuchkyan for useful discussions.

APPENDIX

Let us derive a solution of Eq. (5) for ρ^{rr}_{mn}. We switch to the quantities $F^r_{mn} = \rho^{rr}_{mn}(m!n!)^{1/2}/r!$, for which the equation is

$$\partial F_{mn}{}^r/\partial T = -\frac{r+1}{2}[m(m-1)+n(n-1)]F_{mn}{}^r + F^{r-1}_{m+2,n+2} \tag{A1}$$

with the initial conditions

$$F_{mn}{}^r(0) = f_{mn} = (z^*)^m z^n \exp(-|z|^2)\cdot\delta_{r0}.$$

Taking Laplace transforms, and using the recurrence relations, we find the following expression for the transform of the function F^r_{mn}:

$$\varphi_{mn}{}^r(s) = f_{m+2r,n+2r}\prod_{q=0}^{r}[s+L_{mn}{}^r(q)]^{-1}, \tag{A2}$$

where $L^r_{mn}(q)$ is given in (7). If there are no repeated factors among the $s+L(q)$, $q = 0, 1, \ldots, r$, then the inverse transformation immediately gives us

$$F_{mn}{}^r(T) = f_{m+2r,n+2r}\sum_{q=0}^{r}\exp[-L(q)T]\cdot\prod_{\substack{k=0\\k\neq q}}^{r}[L(k)-L(q)]^{-1}. \tag{A3}$$

The situation becomes more complex, however, if $L(q)$ takes on identical values for different values of q. We will first prove that the number of roots of the equation $L(q) = \text{const}$ is 2 if $q \in [0, r]$. To do this, we plot L^r_{nn} versus q for given values of n and r [for $L^r_{mn}(q)$ with $n = m - 1$, $m - 2$, the assertion can be proved in a corresponding way]. It is convenient to write $L^r_{nn}(q)$ in the form

$$L_{nn}{}^r(q) = (q+1)(N-2q)(N-2q-1),$$

where $N = n + 2r$. For a given value of N, the maximum value of r is obviously $N/2$ or $(N-1)/2$. It follows then from Fig. 5 that the function $L^r_{nn}(q)$ takes on a given value L_0 only at two points q_1 and q_2 which belong to the segment $[0, r_{\max}]$. If there is only one pair of equal factors in the product

$$\prod_{q=0}^{r}(s+L(q))$$

in (A2), then φ^r_{mn} becomes

$$\varphi_{mn}{}^r(s) = f_{m+2r,n+2r}[s+L(q_1)]^{-2}\prod_{\substack{q=0\\q\neq q_1,q_2}}^{r}[s+L(q)]^{-1},$$

and we have, correspondingly,

$$F_{mn}{}^r(T) = f_{m+2r,n+2r}\Bigg\{\sum_{\substack{q=0\\q\neq q_1,q_2}}^{r}\exp[-L(q)T] \times\prod_{\substack{k=0\\k\neq q}}^{r}[L(k)-L(q)]^{-1} + \exp[-L(q_1)T]\cdot\prod_{\substack{k=0\\k\neq q_1,q_2}}^{r}[L(k)-L(q)]^{-1} \times\Bigg[T-\sum_{\substack{q=0\\q\neq q_1,q_2}}^{r}[L(q)-L(q_1)]^{-1}\Bigg]\Bigg\}.$$

Formally, this result can be put in the form

$$F_{mn}{}^{r}(T)=f_{m+2r,n+2r}\left(-\frac{\partial}{\partial L(q_1)}\right)\sum_{\substack{q=0\\ q\neq q_1}}^{r}\exp[-L(q)T]\times\prod_{k=0}^{r}[L(k)-L(q)]^{-1}. \quad (A4)$$

If the number of pairs of equal factors is i, then

$$F_{mn}{}^{r}(T)=f_{m+2r,n+2r}\left[(-1)^{i}\frac{\partial^{i}}{\partial L(q_1)\ldots\partial L(q_i)}\right]\times\sum_{\substack{q=0\\ q\neq q_1',\ldots,q_i'}}^{r}\exp[-L(q)T]\times\prod_{\substack{k=0\\ k\neq q,q_1',\ldots,q_i'}}^{r}[L(k)-L(q)]^{-1}, \quad (A5)$$

where $L(q_i)=L(q_i')$.

Combining (A3)–(A5), we can write the general solution of Eq. (5) in the compact form in (7).

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