Intensity correlation in the cascade laser

Maciej Kolwas and Piotr Lange
Institute of Physics of the Polish Academy of Sciences, Aleja Lotnikow 32/46, 02-668, Warsaw, Poland
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We study the interaction of two optical fields in a laser operating on two coupled lines. A semiclassical model of such a system is developed. We focus our attention on the intensity correlation between the two fields, referred to as cross correlation. This two-photon correlation affects populations of lasing levels as well as the polarization of the medium. A set of field equations derived within this model describes the behavior of the intensities and their correlation along the laser cavity. Numerical solutions of these equations show a substantial increase of the degree of intensity cross correlation in the course of propagation. The cross correlation evolves in a way significantly different from that of the product of intensities. Theoretical results are in good qualitative agreement with the experiment carried out on a helium-neon laser, which is also reported here.

I. INTRODUCTION

Statistical properties of radiation have been a matter of investigation since Planck (1901). Before the laser was invented (1960) the only optical fields available were produced by thermal sources, i.e., they were chaotic in character. The statistical properties of such fields could be characterized completely by the second-order field amplitude correlation function.[1] In the case of nonthermal fields, however, the knowledge of the second-order correlation is not sufficient to describe the statistical properties of a field. Once the first sources of nonthermal optical fields had been invented, the study of higher-order statistical properties of these fields became a new, rapidly developed domain of optics. For a brief survey of this development, see Refs. 2 and 3 and references therein.

The field-correlation function of the order $2n$ (or, alternatively, the photon correlation function of the order n) is directly observable in processes where n-photon absorption (or emission) is involved.[4–6] The rate of n-photon absorption is proportional to the corresponding photon correlation, and not to the product of intensities. The measurements involving photon detection (photon counting, fluorescence intensity, and so on) depend inevitably on the statistics of photons.

As far as one-photon absorption is concerned, one can observe only a first-order photon correlation. In order to observe higher-order correlations, one must seek phenomena where multiphoton processes are important.

This paper is devoted to the study of the second-order photon correlation. The first experiment where the second-order photon correlation was observed, was performed by Hanburry Brown and Twiss[7] in 1958, as yet before the onset of lasers. Since then a number of experiments have been performed, but the unexplored area still remains large. This paper deals with one of the problems recently raised: correlation between photons belonging to two different (but somewhat coupled) light beams of two radically different frequencies. We devote our work to the second-order correlation of photons of different origin, which is often referred to as cross correlation.[8–10] Effects of this kind can be significant when the two fields interact. This is the case of a laser operating on two coupled transitions. Such a device offers a variety of phenomena arising from the interaction of two generated lines. An example is the He-Ne laser operating on two cascading transitions, generating infrared radiation of two wavelengths: 3.39 and 7.69 μm (Fig. 1). It was found that in lasing systems of this kind the cross correlation may be significant and plays an important part in the dynamics of the excitation and populations of the involved atomic levels.[8,9] It occurs in such phenomena as population trapping,[10] optical double resonance,[9] and optical bistability.[11] Up to now most of the works on the cross correlation have studied the problem theoretically; few ideas were tested and confirmed experimentally.

In this paper we present both theory and experiment. This is the continuation of our work, which we reported earlier.[12] We study the interaction between two laser lines generated in cascade, and the role the cross correlation of intensities plays in this interaction. When the correlation is under consideration, it is usually studied or

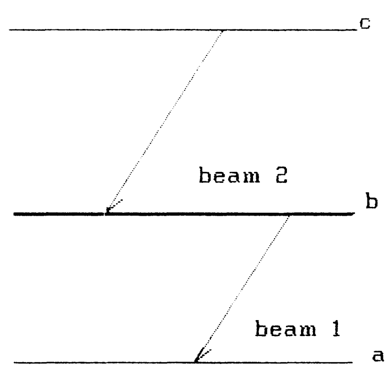

FIG. 1. Scheme of the lasing levels. Two laser beams interacting in cascade.

measured in the time domain using photon counting methods. However, there is another way, namely, its space or position dependence, e.g., along a laser cavity. Field correlations obey wave equations analogous to those for the electric field function $E(\mathbf{r},t)$,[13] where both time and space variables appear. On this analogy we build our model of the time-position evolution of the cross correlation. In Sec. II we develop field equations—for intensities and their cross correlation. In Sec. III we calculate the polarization of a medium along the lines of the semiclassical Lamb theory,[14] extended to the three-level atom by Najmabadi *et al.*[15] In Sec. IV we present and discuss solutions. Section V reports on our experiment, in which we observed phenomena discussed in Secs. II–IV. Comparison of theory with experiment and conclusions follow in Secs. V and VI.

II. FIELD EQUATIONS

Our treatment is based on the classical representation of the radiation field by c-number amplitudes $\mathcal{E}_1$ and $\mathcal{E}_2$ for two modes with frequencies ν_1 and ν_2, respectively:

$$\mathcal{E}_i = E_i^+(\mathbf{r},t)\exp[i(\nu_i t - k_i z)] + E_i^-(\mathbf{r},t)\exp[i(\nu_i t + k_i z)] + \text{c.c.}, \quad i=1,2 . \tag{1}$$

We distinguish forward- and backward-running wave amplitudes E_i^+, E_i^-. Unlike in Lamb[14] or Najmabadi[15] theory, E_i^- and E_i^+ are generally random functions of time and position. They are subject to random fluctuations arising from thermal motion, mechanical vibrations, and many other effects. Our objective is to derive equations of motion for stochastically averaged intensities $\langle I_1\rangle, \langle I_2\rangle$, as well as for their cross correlation. Although we consider random variables, we shall not delve into the probability density $P(I_1,I_2,t)$ to know details of photon correlations. Instead, we assume their existence and we want to study their evolution. Correlations (the cross correlation $\langle I_1 I_2\rangle$, too) obey wave equations analogous to those for the amplitudes.[6,13] We derive our basic equations along the lines of Lamb's theory (see, e.g., Ref. 14), assuming a slowly varying envelope approximation (SVEA), i.e.,

$$\left|\frac{\partial^2 E_i}{\partial z^2} k_i\right| \ll \left|\frac{\partial E_i}{\partial z}\right| .$$

We consider only the longitudinal variation of the amplitudes (along the z axis, parallel to the resonator), neglecting their transverse variation. Under these approximations one obtains the following equations (for details, see, e.g. Ref. 14):

$$\frac{\partial E_i^\pm}{\partial t} + c\frac{\partial E_i^\pm}{\partial t} = -\kappa_i E_i^\pm + \frac{i\nu_i}{2\epsilon}P_i^\pm , \tag{2}$$

where $P_i^\pm$ is the Fourier component of the polarization corresponding to the frequency ν_i and the wave vector k_i, and ϵ is the permitivity of medium. The damping parameters κ_i account for electromagnetic energy dissipation. In the Lamb theory[14] all losses are modeled by a finite conductivity of the medium, so κ is related to it: $\kappa=\sigma/(2\epsilon)$.

Equation (2) is the basis from which one can develop equations for any product of $E_i^\pm$, their complex conjugate, and then for their averages (for example, see Ref. 13, p. 51). The radiation fields interact resonantly with atoms: $\mathcal{E}_1$ with the lower transition, and $\mathcal{E}_2$ with the upper transition (see Fig. 1). They induce atomic dipole moments, which in turn act on the fields, because the polarizations P_i of Eq. (2) are the mean atomic dipole moments in a unit volume of the proper frequency. P_i's are expressed in terms of atomic density matrix elements:

$$P_1 = \sigma_{ab} D_{ba} + \text{c.c.} , \tag{3a}$$

$$P_2 = \sigma_{bc} D_{cb} + \text{c.c.} , \tag{3b}$$

where σ_{ab} is the slowly varying part of the density-matrix element ρ_{ab} (which rotates with the frequency ν_1), and likewise σ_{bc} is the slowly varying part of ρ_{bc}; D_{ba}, D_{cb} are the electric dipole matrix elements. It is convenient to work with quantities $V_1 = E_1 D_{ab}/i\hbar$, $V_2 = E_2 D_{bc}/i\hbar$, which we will use from now on. Starting with Eq. (2) one obtains evolution equations for products which are subsequently formally averaged. In this way from double products one obtains observable intensities:

$$\left[\frac{\partial}{\partial t} \pm \frac{\partial}{\partial z}\right]\langle V_1^\pm (V_1^\pm)^*\rangle = -2\kappa_1\langle V_1^\pm (V_1^\pm)^*\rangle - G_1[\langle (V_1^\pm)^*\sigma_{ab}^\pm\rangle + \text{c.c.}] , \tag{4a}$$

$$\left[\frac{\partial}{\partial t} \pm \frac{\partial}{\partial z}\right]\langle V_2^\pm (V_2^\pm)^*\rangle = -2\kappa_2\langle V_2^\pm (V_2^\pm)^*\rangle - G_2[\langle (V_2^\pm)^*\sigma_{bc}^\pm\rangle + \text{c.c.}] , \tag{4b}$$

$G_1 = \nu_1|D_{ab}|^2/(2\epsilon\hbar)$, $G_2 = \nu_2|D_{bc}|^2/(2\epsilon\hbar)$, while from fourfold products the cross correlation

$$\begin{aligned}\left[\frac{\partial}{\partial t} \pm \frac{\partial}{\partial z}\right]\langle I_1^\pm I_2^\pm\rangle &= -2(\kappa_1+\kappa_2)\langle I_1^\pm I_2^\pm\rangle \\ &\quad - G_1[\langle V_2^- (V_2^-)^* (V_1^-)^* \sigma_{ab}^-\rangle + \text{c.c.}] \\ &\quad - G_2[\langle V_1^\pm (V_1^\pm)^* (V_2^\pm)^* \sigma_{bc}^\pm\rangle + \text{c.c.}] .\end{aligned} \tag{4c}$$

Here $\sigma^\pm$ denotes the Fourier component of σ (decomposition with respect to wave vectors) having the wave vector $\pm k_1$ in the case of σ_{ab} and $\pm k_2$ in the case of σ_{bc}. Such components appear explicitly in most treatments in a natural way without any extra effort.

From Eq. (4) one can see the influence of atoms on the radiation fields. To see how the radiation affects the atoms, we solve the density-matrix equations of evolution. Apart from the coherencies σ_{ab}, σ_{bc} we will need the populations ρ_b, ρ_c, which contain information about field quantities: intensities and intensity correlation, as we explain in Sec. IV.

III. ATOMIC DENSITY MATRIX

We describe the atomic medium by a density matrix ρ_{mn}, $m,n=a,b,c$. Under the rotating wave approximation its elements obey the equations

$$\frac{d\rho_a}{dt}=\Lambda_a-\gamma_a\rho_a-(V_1^*\sigma_{ab}-\text{c.c.})\ , \tag{5a}$$

$$\frac{d\rho_b}{dt}=\Lambda_b-\gamma_b\rho_b-(V_2^*\sigma_{bc}+\text{c.c.})+(V_1^*\sigma_{ab}+\text{c.c.})\ , \tag{5b}$$

$$\frac{d\rho_c}{dt}=\Lambda_c-\gamma_c\rho_c+(V_2^*\sigma_{bc}+\text{c.c.})\ , \tag{5c}$$

$$\frac{d\sigma_{ab}}{dt}=-z_{ab}\sigma_{ab}-(\rho_b-\rho_a)V_1^*-V_2\sigma_{ac}\ , \tag{5d}$$

$$\frac{d\sigma_{bc}}{dt}=-z_{bc}\sigma_{bc}-(\rho_c-\rho_b)V_2+V_1^*\sigma_{ac}\ , \tag{5e}$$

$$\frac{d\sigma_{ac}}{dt}=-z_{ac}\sigma_{ac}+V_2\sigma_{ab}-V_1\sigma_{bc}\ , \tag{5f}$$

plus relations

$$\sigma_{mn}=\sigma_{nm}^*,\quad \rho_{ab}=\sigma_{ab}\exp(i\nu_1 t)\ ,$$
$$\rho_{bc}=\sigma_{bc}\exp(i\nu_2 t),\quad \rho_{ac}=\sigma_{ac}\exp[i(\nu_1+\nu_2)t]\ ;$$

$V_{1,2}$ stands for $V_{1,2}^-+V_{1,2}^+$; Λ's represent incoherent pumping of the lasing levels, i.e., excitation via He-Ne collisions; and z's combine damping and detuning:

$$z_{mn}=\gamma_{mn}+i\Delta_{mn},\quad \Delta_{ab}=\omega_b-\omega_a-\nu_1\ ,$$
$$\Delta_{bc}=\omega_c-\omega_b-\nu_2,\quad \Delta_{ac}=\Delta_{ab}+\Delta_{bc}\ .$$

In the case of gaseous media the movement of atoms must be taken into account. It amounts to the replacement of kz by $kz+vt$, where v is the z component of the atomic velocity. It means that Eqs. (5) are to be solved for each velocity interval $(v,v+dv)$ and the final matrix is velocity averaged.

We apply a perturbative approach up to fourth order, i.e., to the lowest order the product of two fields appears in populations. Validity of the perturbation approach depends on the ratio (Rabi frequency)/(damping rate of the system), which should be smaller than 1. The Rabi frequency $D_{mn}E/\hbar$ is determined and may be controlled by the intensity (which can be handled by the pumping rate). The damping rate is determined for the most part by three phenomena: (i) spontaneous emission, (ii) collisions; and (iii) laser fluctuations. If the laser line is broad (e.g., on account of Doppler broadening), the correlation time is short and the damping rate is dominated by a large linewidth. In this case the light fluctuations affect strongly the light-atom interaction and improve conditions for the validity of the perturbation approach. Keeping in mind this role of light fluctuations, we assume the condition we have discussed above to be satisfied, permitting us to use the perturbation approach. Note that our field quantities are random variables; therefore we cannot assume that they vary little during a typical lifetime, the assumption being widely used.[14,15]

We have used the slowly varying envelope approximation to the random-field amplitudes. However, inasmuch as we work with average quantities quadratic in amplitude, we actually assume only these quantities to be slowly varying. We could start with a wave equation directly for correlations,[6] arriving at Eq. (4), leaving out Eq. (2). Besides, the assumption that E is slowly varying involves that it varies little over a lapse of time of the order 10^{-14} s, which is much shorter than a typical atomic relaxation time (10^{-8}–10^{-7} s). The random character of a field does not determine the time scale of its random changes. It is a question of further assumptions, and there are two different approaches to this problem (Ref. 16, p. 194).

Now let us write formal solutions for the third-order polarizations:

$$\begin{aligned}V^*(t)\sigma+\text{c.c.}=&-N_{ba}\int_0^t dt' g_1(t,t')\\&+N_{ba}\int_0^t dt' g_1(t,t')\int_0^{t'} dt''(e^{\gamma_a(t''-t')}+e^{\gamma_b(t''-t')})\int_0^{t''} dt''' g_1(t'',t''')\\&-N_{cb}\int_0^t dt' g_1(t,t')\int_0^{t'} dt'' e^{\gamma_b(t''-t')}\int_0^{t''} dt''' g_2(t'',t''')\\&+N_{ba}\int_0^t dt'\int_0^{t'} dt''\int_0^{t''} dt''' g(t,t',t'',t''')e^{z_{ab}(t'-t)+z_{ac}(t''-t')+z_{ab}(t'''-t'')}\\&-N_{cb}\int_0^t dt'\int_0^{t'} dt''\int_0^{t''} dt''' g(t,t',t''',t'')e^{z_{ab}(t'-t)+z_{ac}(t''-t')+z_{bc}(t'''-t'')}+\text{c.c.}\ ,\end{aligned} \tag{6a}$$

$$\begin{aligned}V_2^*(t)\sigma_{bc}+\text{c.c.}=&-N_{cb}\int_0^t dt' g_2(t,t')\\&+N_{cb}\int_0^t dt' g_2(t,t')\int_0^{t'} dt''(e^{\gamma_b(t''-t')}+e^{\gamma_c(t''-t')})\int_0^{t''} dt''' g_2(t'',t''')\\&-N_{ba}\int_0^t dt' g_2(t,t')\int_0^{t'} dt'' e^{\gamma_b(t''-t')}\int_0^{t''} dt''' g_1(t'',t''')\\&+N_{cb}\int_0^t dt'\int_0^{t'} dt''\int_0^{t''} dt''' g(t',t,t'',t''')e^{z_{bc}(t'-t)+z_{ac}(t''-t')+z_{bc}(t'''-t'')}\\&-N_{ba}\int_0^t dt'\int_0^{t'} dt''\int_0^{t''} dt''' g(t',t,t''',t'')e^{z_{bc}(t'-t)+z_{ac}(t''-t')+z_{ab}(t'''-t'')}+\text{c.c.}\ ,\end{aligned} \tag{6b}$$

with the notation

$$g_1(t,t')=\tfrac{1}{2}[V_1^*(t)V_1(t')e^{z_{ab}(t'-t)}+\text{c.c.}]\ ,$$

$$g_2(t,t')=\tfrac{1}{2}[V_2^*(t)V_2(t')e^{z_{bc}(t'-t)}+\text{c.c.}]\ ;$$

$$g(t,t',t'',t''')=V_1^*(t)V_2^*(t')V_1(t'')V_2(t''')\ ;$$

$$N_{ij}=\Lambda_i/\gamma_i-\Lambda_j/\gamma_j\ .$$

The expressions (6a) and (6b) will serve for evaluation of the right-hand sides of Eqs. (4a)–(4c). For brevity we have not written out explicitly the V^+ and V^- amplitudes. To obtain the definite form of a particular equation (for the + or − wave) one must put $(V^+)^*(t)$ or $(V^-)^*(t)$ instead of $V^*(t)$ [which stands for the sum $(V^+)^*+(V^-)^*$] and pull out of Eq. (6a) or (6b) the part in which the exponentials e^{kz} cancel themselves. The polarizations (6a) and (6b) must be averaged. With a view to performing it, we note that fourth-order correlations will appear only in the last two integrals. They represent coherent transitions.[16] They come from the double quantum coherence σ_{ac}. The remaining fourfold products (second and third terms) come from population differences; they are due to stepwise transitions involving the intermediate level. The amplitudes V_1 and V_2 appearing there cannot be significantly correlated because they are separated by the time interval $(t'-t'')$, which is of the order of b-state lifetime. In these terms there will be just a product of intensities, i.e., only one-photon correlations.

All in all, the polarizations consist of four parts [listed in the order in which they appear in the formulas (6a) and (6b)]: (i) linear gain, dependent on the second-order autocorrelation (i.e., intensity); (ii) nonlinear saturation, dependent on the square of intensity; (iii) nonlinear gain, dependent on the both intensities; and (iv) coherent terms, dependent on the cross-correlation. We shall return to the physical discussion of these terms in subsequent sections.

Now we face the problem of evaluating the integrals in (6). The strict evaluation is beyond the scope of this paper, but as we argue below, it is not necessary to our purpose. Nevertheless, we must know something about the time behavior of the integrands in (6). The first assumption is the steady state of the stochastic quantities that will appear while averaging Eq. (6). This assumption is very often encountered in the literature[1,17] and physically signifies that the results of an observation are independent of the moment the observation starts. Under further assumptions determining the nature of a random process, one obtains the time dependence of a correlation function. For example, in the case of the second-order correlation of a Gaussian process, one obtains exponential decay of the type $\exp[-\gamma|t_1-t_2|]$.[17] If we put

$$\langle V^*(t)V(t')\rangle=\langle V^*(t)V(t)\rangle\exp[\gamma(t'-t)]$$

we see that the time decay of the field correlation results in augmenting an atomic relaxation rate. In the case of Gaussian processes, higher-order even correlations can be expressed as products of the second-order correlation.[17] We do not assume our process to be Gaussian because it is not the case of the system we describe. We assume only that a multi-time-correlation can be expressed by an analogous one-time-correlation multiplied by a function of time independent of the fields:

$$\langle V_i^*(t)V_i(t)\rangle=\langle V_i^*(t)V_i(t)\rangle f_i(t',t'')$$
$$=\langle I_i\rangle f_i(t',t'')\ , \tag{7a}$$

$$\langle V_i^*(t)V_j^*(t')V_i(t'')V_j(t''')\rangle$$
$$=\langle I_iI_j\rangle f_{ij}(t',t'',t''')\ , \tag{7b}$$

$$\langle V_i^*(t)V_j^*(t)V_k^*(t')V_i(t)V_j(t'')V_k(t''')\rangle$$
$$=\langle I_iI_jI_k\rangle f_{ijk}(t',t'',t''')\ . \tag{7c}$$

This is the case in all typical models met in the literature.[17] In this way we can pull the fields out of integrals and represent an integral by a coefficient dependent on atomic constants and on the functions f_i, f_{ij}, and f_{ijk}, but independent of the field amplitudes. As we mentioned above, we do not need exact values of these coefficients. The only thing we can say, and which is essential, is that the functions f_i, f_{ij}, and f_{ijk} introduce an additional damping, which is the greater, the greater the order of the correlation. As a result, the coefficients of Eq. (4c) will be smaller than they would be in the equation for the product $\langle I_1\rangle\langle I_2\rangle$.

In Eq. (6) we have also three-photon correlations. As we do not go beyond two-photon phenomena, we are forced to factorize them (with a change of the coefficient appearing there), in order that Eq. (6) form a closed set. Finally, we arrive at the following equations:

$$\frac{\partial \mathcal{I}_1^+}{\partial z}=a_1\mathcal{I}_1^+-b_1(\mathcal{I}_1^+)^2-c_1\mathcal{I}_1^+\mathcal{I}_1^-+d_1^+\mathcal{I}_1^+\mathcal{I}_2+e_1^+\mathcal{Y}^+\ , \tag{8a}$$

$$-\frac{\partial \mathcal{I}_1^-}{\partial z}=a_1\mathcal{I}_1^--b_1(\mathcal{I}_1^-)^2-c_1\mathcal{I}_1^+\mathcal{I}_1^-$$
$$+d_1^-\mathcal{I}_1^-\mathcal{I}_2+e_1^-\mathcal{Y}^-\ , \tag{8b}$$

$$\frac{\partial \mathcal{I}_2}{\partial z}=a_2\mathcal{I}_2-b_2\mathcal{I}_2^2+d_2^+\mathcal{I}_2\mathcal{I}_1^++d_2^-\mathcal{I}_2\mathcal{I}_1^-$$
$$-e_2^-\mathcal{Y}^--e_2^+\mathcal{Y}^+\ , \tag{8c}$$

$$\frac{\partial \mathcal{Y}^+}{\partial z}=(\alpha^+-\beta^+\mathcal{I}_1^+-\beta^-\mathcal{I}_1^--\delta^+\mathcal{I}_2)\mathcal{Y}^++\epsilon^+\mathcal{I}_1^+\mathcal{I}_2^2\ , \tag{8d}$$

$$\frac{\partial \mathcal{Y}^-}{\partial z}=(\alpha^-+\gamma^+\mathcal{I}_1^++\gamma^-\mathcal{I}_1^--\delta^-\mathcal{I}_2)\mathcal{Y}^-+\epsilon^-\mathcal{I}_1^-\mathcal{I}_2^2\ , \tag{8e}$$

where the $\mathcal{I}$'s are the averaged intensities and the $\mathcal{Y}$'s are the cross correlations. The superscripts ± with $\mathcal{I}_2$ are missing because we will consider only one wave of this frequency, i.e., it will be a traveling wave.

Time derivatives have been omitted, because we study a steady-state regime. All the parameters are positive except for the $e_{1,2}^{\pm}$'s, which have the form $\alpha N_{ba}-\beta N_{cb}$.

The sign is difficult to determine if all three levels are excited incoherently. If one wants to keep track of a definite transition path, one must start with a definite zero-field population, putting zero for the two remaining ones. In this case the signs of the two terms representing two-photon transitions are the same, as it should be. Anticipating solutions, we note that general features do not depend on the sign of this term, since this term is the smallest of all.

IV. BEHAVIOR OF THE FIELD QUANTITIES

Equations (8) describe the position dependence of the field quantities: intensities and cross correlations. The parameters come from and correspond to the respective terms of the polarizations (6): a refers to the linear buildup; b and c are the self-saturation and competition terms, respectively; d is the interaction between beams 1 and 2 via two-step two-photon transitions; and e is the interaction between beams 1 and 2 via coherent two-photon transitions.

We have to solve a set of ordinary differential equations. Exact solution seems to be impossible in the general case. Even if we could obtain general formulas, they would be vague and difficult to discuss because of a number of parameters. Even rough approximations lead to complex formulas, and their numerical analysis is necessary. For this reason we prefer to present results of numerical simulation. We solved Eq. (8) numerically, imposing mathematical conditions that expressed specific physical situations: (i) free propagation of both beams; and (ii) beam 1 is a closed resonator, whereas beam 2 is a running wave.

Before the numerical computation, it is worthwhile to examine the order of magnitude of the entering parameters. It is convenient to work with dimensionless quantities: $V_1^* V_1$ divided by $(\gamma_a \gamma_b / \gamma_{ab})^2$ and $V_2^* V_2$ by $(\gamma_b \gamma_c / \gamma_{bc})^2$. If the γ's are measured in megahertz, then the dimensionless intensities should be less than 1, and the linear gain can take on values of 1–10. The range of values of the nonlinear coefficients depends on the actual scaling of the intensities within the range 0–1. Roughly speaking, the greater the intensities, the smaller the coefficients.

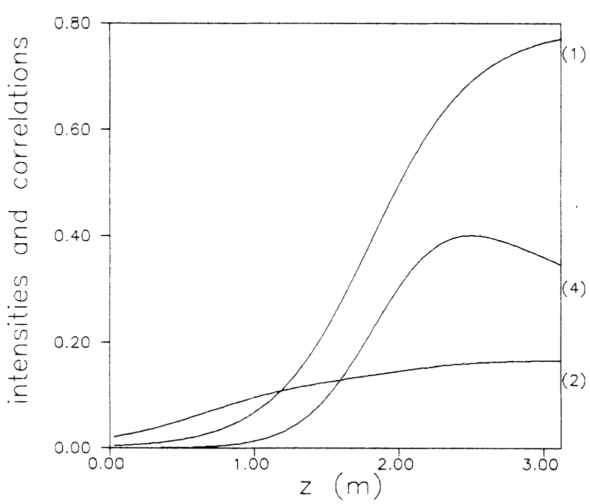

FIG. 2. Propagation of dimensionless intensities and their correlation through an active (amplifying) medium: solutions to Eqs. (8) (which in this case are reduced to three equations). The labels throughout all figures denote (1) $\mathcal{I}_2$, (2) $\mathcal{I}_1^+$, (3) $\mathcal{I}_1^-$, (4) $\mathcal{Y}^+$, and (5) $\mathcal{Y}^-$. The parameters are in the Appendix.

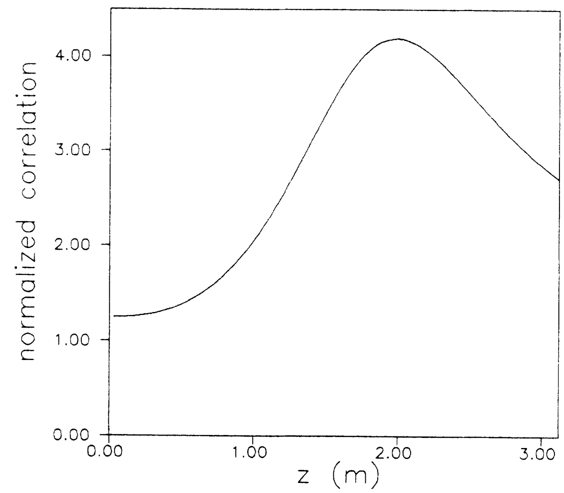

FIG. 3. Degree of correlation $\mathcal{Y}^+/(\mathcal{I}_1^+ \mathcal{I}_2)$ in the course of propagation of Fig. 2. The rise is over three times, and a falloff to a steady state (attained beyond the plot) follows.

The coefficients b and d arise from the second and the third term of (6), respectively. One can see the main difference between them: The former has the sum

$$\exp[\gamma_a(t''-t')]+\exp[\gamma_b(t''-t')] ,$$

whereas the latter has only $\exp[\gamma_b(t''-t')]$. Thus we

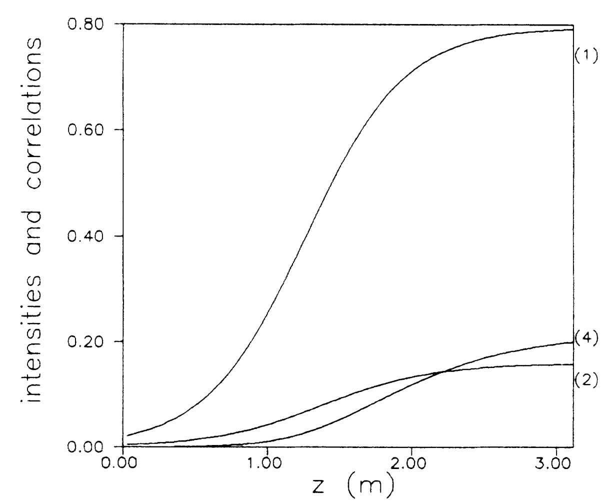

FIG. 4. As Fig. 2, but with interchanged initial values.

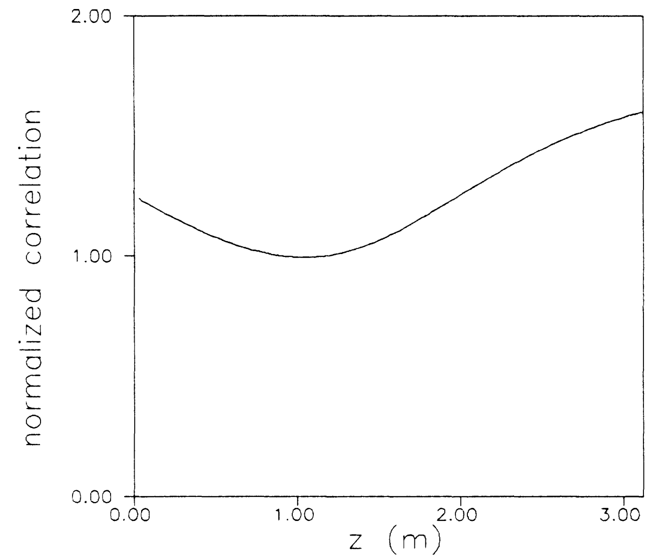

FIG. 5. The degree of correlation—case of Fig. 3; advanced $\mathcal{I}_2$ can lead to a transient anticorrelation.

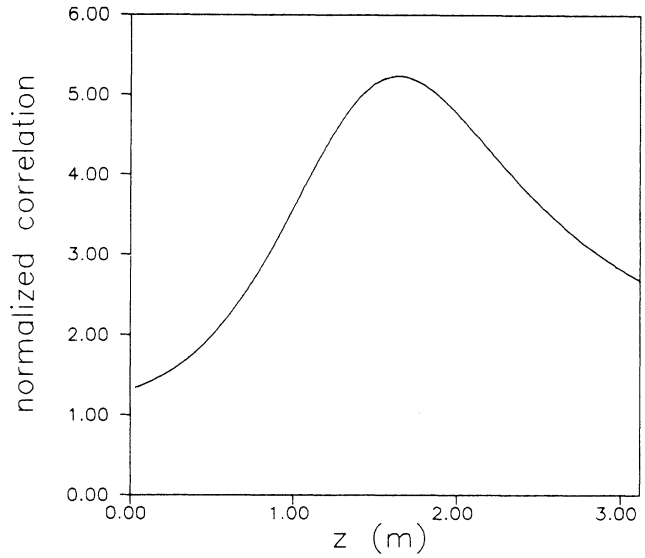

FIG. 7. Degree of correlation for the case of Fig. 6. The rise is over four times.

should expect c greater than d. If the b state has a lifetime short compared to the lifetimes of the a and c states (condition for a large gain of the pumping beam 2), then d is considerably smaller than c. Relative magnitudes of coefficients are discussed in greater detail in Ref. 15.

The parameters are modified, compared to the analogs of Najmabadi,[15] by laser fluctuations and the factorization procedure carried out in the cross-correlation equations. The precise extent of this modification is beyond the scope of our theory. We examined a number of solutions with various sets of parameters, which we chose along the lines of the above discussion. The main features of the solutions, which we emphasize below, can be obtained with coefficients within a fairly large range. We present the results in Figs. 2–11 for a few samples of parameters.

A. Free propagation

If we suppress $\mathcal{I}_1^-$ and $\mathcal{Y}^-$, we have the free propagation of two traveling waves and of their cross correlation through an active medium. The situation is shown in

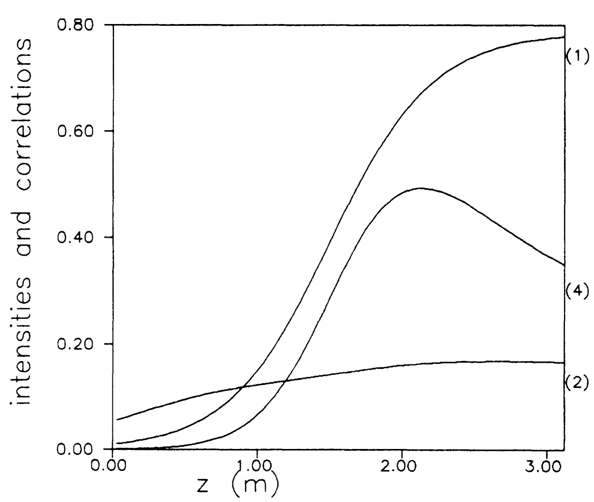

FIG. 6. As Fig. 2, with one more set of initial values. Here both beams are fairly intense, having initial values equal to those of the next case (closed resonator).

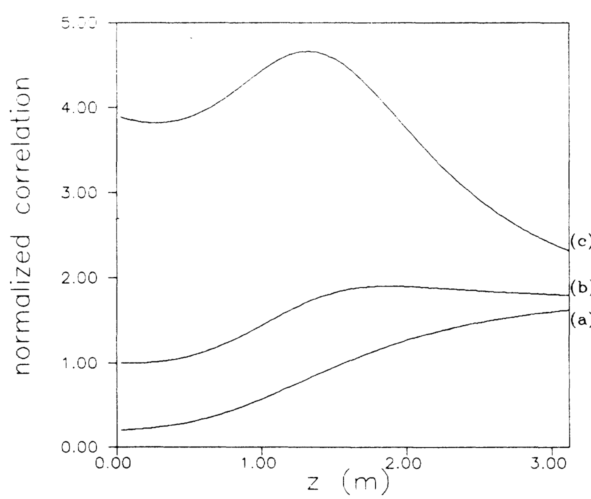

FIG. 8. Degree of correlation—three other cases: (a) a rise from a small value 0.2, (b) a smaller rise from the initial value 1, and (c) a falloff for too large an initial value. The degree of cross correlation tends to keep a "reasonable value."

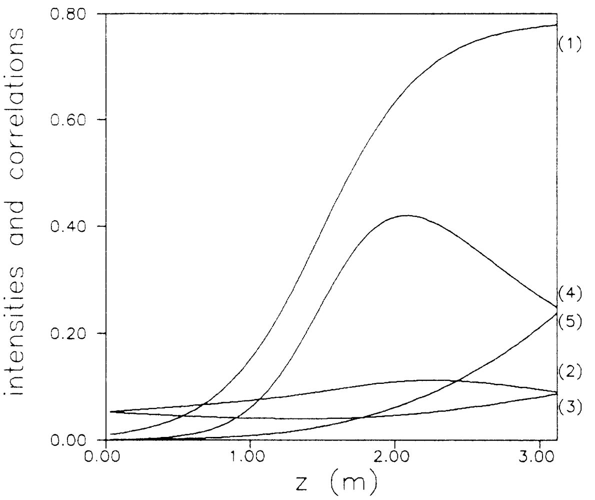

FIG. 9. Closed resonator (for beam 1 only). Coefficients and initial values for $\mathcal{I}_1^+$, $\mathcal{Y}^+$, and $\mathcal{I}_2$ are the same as in Fig. 6; the labels are the same as in Fig. 2. The influence of the counterpropagating beam on the degree of correlation is of minor importance (compare Fig. 7).

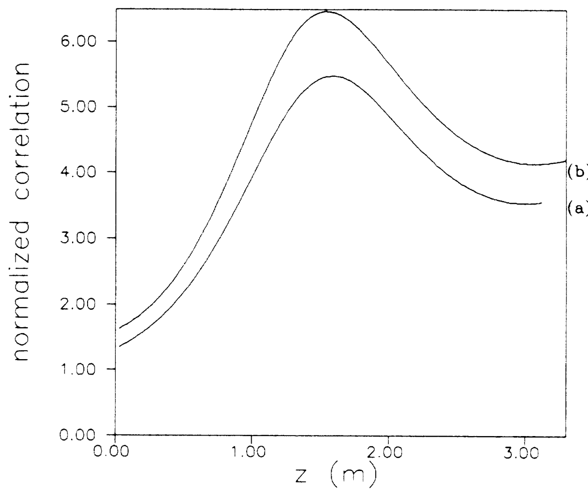

FIG. 11. Degree of correlation: (a) case of Fig. 9, and (b) case of Fig. 10 (other parameters and a little longer resonator).

Figs. 2–8 for a sample of coefficients and different boundary values. Evolution of the cross correlation depends essentially upon relative boundary values; for certain values the normalized cross correlation (the degree of cross correlation $\langle I_1\rangle\langle I_2\rangle/\langle I_1 I_2\rangle$) does not change appreciably [Figs. 5, 8(b), and 8(c)], but for others it exhibits a considerable growth and a maximum occurs [Figs. 3, 6, and 8(a)]. It is clearly seen that the degree of cross correlation cannot rise excessively, neither can it fall too low. The character of its proceeding behavior depends on (i) the history (preceding evolution), and (ii) the relative values of the intensities making up the correlation. Generally, an asymmetry between the two beams is needed for the correlation to increase substantially. It can be understood with the help of a picture of two cascading waves: they help each other; the stronger precipitates the weaker. The sensitivity of the correlation to relative values of the intensities is easily seen in Figs. 2–5.

All the quantities attain the "the steady state," i.e., they get saturated. The role of the coupling in the propagation can be seen by the comparison of the saturated values of intensities with those in the absence of the coupling (which is given by the ratio a/b). The influence of one beam on another depends more strongly on the coupling coefficient e coming from the coherent transitions than on d, i.e., the one due to the stepwise transitions. Beam 1 is much more sensitive to the coupling because it is weaker relative to beam 2 and, moreover, the two coupling coefficients have the same, positive sign [Eq. (8a)]. It is more evident in the second case, where the resonator is closed for beam 1. The sensitivity to this kind of coupling results from the rapid growth of the cross correlation with the propagation of the beams.

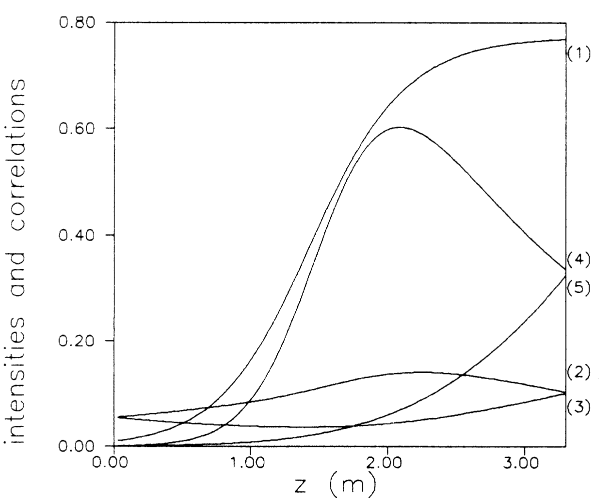

FIG. 10. As in Fig. 8 but another set of parameters (in the Appendix). Stronger coupling between beams 1 and 2 results in a greater sum of the + and − beam 1; their difference is greater too.

B. One of the beams in closed resonator

Figures 9–11 show the case of the closed resonator for beam 1, while beam 2 is still a traveling wave. The simplest way to realize such a situation is reported in Sec. V. This case is more interesting because it represents intracavity conditions and describes the situation of our experiment. Here again we have the propagation of beam 2, which brings about an interesting behavior of the cross correlation, distinctly different from that of the intensity product. It should be emphasized that this effect is again obtained by propagation of the beams. The parameters are such that $\mathcal{I}_2$ grows rapidly and is finally much more intense than $\mathcal{I}_1$. Under these conditions the coupling with $\mathcal{I}_1$ has little effect on $\mathcal{I}_2$. The other beam, $\mathcal{I}_1$ is much more sensitive to the coupling with $\mathcal{I}_2$. Owing to this coupling its value at the right boundary is fairly higher than at the left end of the resonator, where $\mathcal{I}_2$ sets off. The right-hand mirror level of $\mathcal{I}_1$ can be handled by the e parameter as it was in the prior case, but now it is even more evident. Note that the overall power of beam 1, divided into two parts is approximately equal to the intensity of the single, "one-way" wave of the prior case.

The cross correlation $\mathcal{Y}^+$ starts with a value greater than $\mathcal{I}_1^+(0)\mathcal{I}_2(0)$, grows fast along the resonator, faster than the product $\mathcal{I}_1^+\mathcal{I}_2$, then at the right-hand mirror it becomes $\mathcal{Y}^-$—its counterpropagating counterpart which relaxes throughout the cavity to the initial value of $\mathcal{Y}^+$. The main result of this computation is shown in Fig. 11. This is the degree of correlation $\langle \mathcal{I}_1\mathcal{I}_2\rangle/\langle \mathcal{I}_1\rangle\langle \mathcal{I}_2\rangle$ ($\mathcal{I}_1$ stands for the sum $\mathcal{I}_1^+ + \mathcal{I}_1^-$). This quantity varies along the cavity. It forms a fairly high peak roughly half-way from the left-hand mirror to the right-hand one. The reason for this is that the light fluctuations modify coupling strengths; although, on account of these modifications, the linear gain is less than the sum of the corresponding coefficients for $\mathcal{I}_1$ and $\mathcal{I}_2$, but the nonlinear terms become diminished too. These modifications result in a retardation of the saturation. It manifests itself most distinctly if one of the beams is a traveling wave. The peak does not result from the superposition of $\mathcal{Y}^+$ and $\mathcal{Y}^-$; although we have depicted the combined normalized correlation, the effect remains in the quantity $\mathcal{Y}^+/(\mathcal{I}_1^+\mathcal{I}_2)$. The other correlation $\mathcal{Y}^-$ is less significant, again because of the asymmetry introduced by the unidirectional beam 2. In the case of $\mathcal{Y}(0) < \mathcal{I}_1(0)\mathcal{I}_2(0)$ the patterns are only slightly modified; in fact, from the mathematical standpoint this change is a matter of rescaling. On the other hand, from the physical point of view it can be a different case.

The computational results presented here were observed experimentally in a He-Ne laser operating on the lines 3.39 μm and 7.69 μm. As we stated earlier, the ranges of possible values of parameters is fairly large but they are not independent of each other if we seek nonexploding solutions. Once the linear coefficients have been fixed, the others are confined to quite a narrow range. We present the results for two sets of coefficients, but one can reproduce (qualitatively) the results for other, significantly different sets. It is important, for we cannot specify their values corresponding to a particular experiment. The parameters we chose do not necessarily match those of our experiment.

V. EXPERIMENT

A. Principles of measurement

The polarizations calculated in Sec. II, put into the density-matrix equation, yield fourth-order populations. Following the procedure discussed in Sec. II, the averaged populations are

$$\rho_a = \rho_a^{(0)} + A_1\langle I_1\rangle - B_1\langle I_1\rangle^2 - C_1\langle I_1\rangle\langle I_2\rangle + D_1\langle I_1 I_2\rangle \, , \tag{9a}$$

$$\rho_c = \rho_c^{(0)} - A_2\langle I_2\rangle + B_2\langle I_2\rangle^2 - C_2\langle I_1\rangle\langle I_2\rangle - D_2\langle I_1 I_2\rangle \, . \tag{9b}$$

The letters A,B,C,D denote constants that are independent of the average field quantities. They are positive. The terms with both intensities represent two photon transitions: absorption or emission of one photon from either beam. The net rate of the transitions is not zero because of relaxation processes. There are two types of two-photon absorption: stepwise and coherent. The former, involving an intermediate atomic-level population, depends on the product of intensities; the latter is due to two-photon coherence and depends on the cross correlation.

The populations can be observed by means of fluorescence measurements. Fluorescence from a given level is proportional to the population of this level, which is affected by radiation-field quantities: intensities and correlations (auto-correlations and cross correlations). Thus fluorescence measurement contains information about these quantities.

The dominant terms in (9) are of course the linear ones. If the whole population is observed, i.e., if the linear term is not cut out by an appropriate observation technique, one can regard the population as proportional to the intensity, leaving out of account the nonlinear part. In order to select "two-photon" terms, the "one-photon" ones must be eliminated (along with the field-independent $\rho^{(0)}$). This is feasible by a measurement of the fluorescence from a given level by a lock-in detector with a proper light beam chopped. For example, if beam 1 is chopped, the lock-in detector being tuned to the chopping frequency and a monochromator tuned to a fluorescence line from level c, then only the part of ρ_c dependent on beam 1 will be detected, whereas the first three terms of (9.2) will be cut out of detection. It was actually one of the signals we measured. We denote it S_1. In the lowest order of approximation it is proportional to the bilinear terms of (9.2). If in turn the whole fluorescence is measured, then, as we argued earlier, the signal can be thought of as proportional to the intensity $\langle I_2\rangle$ (but one must remember the constant $\rho_c^{(0)}$). This was our second signal S_2. Similarly, the fluorescence intensity from level a gives a signal S_3 proportional to $\langle I_1\rangle$. Let us write out our three signals:

$$S_1 \propto C_2\langle I_1\rangle\langle I_2\rangle + D_2\langle I_1 I_2\rangle \, , \tag{10a}$$

$$S_2 \propto -\langle I_2\rangle \, , \tag{10b}$$

$$S_3 \propto +\langle I_1\rangle \, , \tag{10c}$$

where $\propto$ stands for "proportional to."

Once the three signals have been detected, one can construct a degree of cross correlation:

$$\mathcal{A} = S_1/(S_2 S_3) = A + B\langle I_1 I_2\rangle/(\langle I_1\rangle\langle I_2\rangle) \, , \tag{11}$$

where A and B are constants independent of intensities. This is a function of position in a laser discharge tube, z coordinate.

If the character of the z dependence of the cross corre-

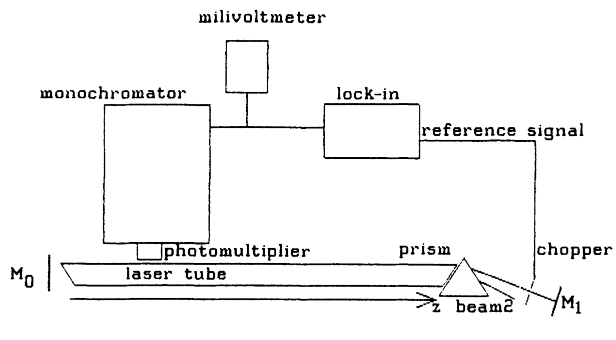

FIG. 12. Experimental setup.

lation is different from that of the product of the intensities, it will be reflected in the degree of cross correlation $\mathcal{A}$. This is the principle of our detection and measurement of the degree of cross correlation. We measured the signals S_1, S_2, and S_3 as functions of z and constructed $\mathcal{A}(z)$.

B. Experimental setup

Our arrangement is shown in Fig. 12. A homemade He-Ne laser was used, with a discharge tube 2.77 m long, operating on two coupled lines: 7.69 μm (b_1), lower transition; and 3.39 μm (b_2), upper transition. The latter is much stronger and plays the part of a pump for the former. The beams were split spatially inside the resonator with a prism (sodium fluoride). The splitting of the beams served two aims: (i) to make b_2 a traveling wave without any perturbation of b_1, and (ii) to chop the beam b_1 only. To make b_2 a traveling wave, it sufficed to let it become reflected only on the mirror M_0, the second one

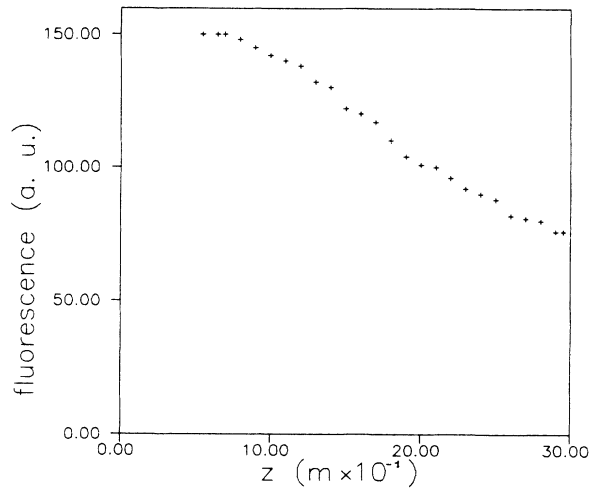

FIG. 14. Signal S_2 of (10.2) vs z coordinate.

missing, because the generation of b_2 is very strong and a double passage propagation (towards—and from—the mirror M_0) is sufficient for its buildup.

We observed the fluorescence emitted perpendicularly to the laser tube on two lines: 543.4 nm from level c, and 794.3 nm from level a, as a function of position. Measurements were made at 25 points, each 10 cm, moving the monochromator mounted on a special carrier. The direct light detector—photomultiplier EMI 9558QA—remained the same for all the measurements.

C. Results and discussion

We carried out the measurements in the situation described above: b_1, standing wave; and b_2 running wave.

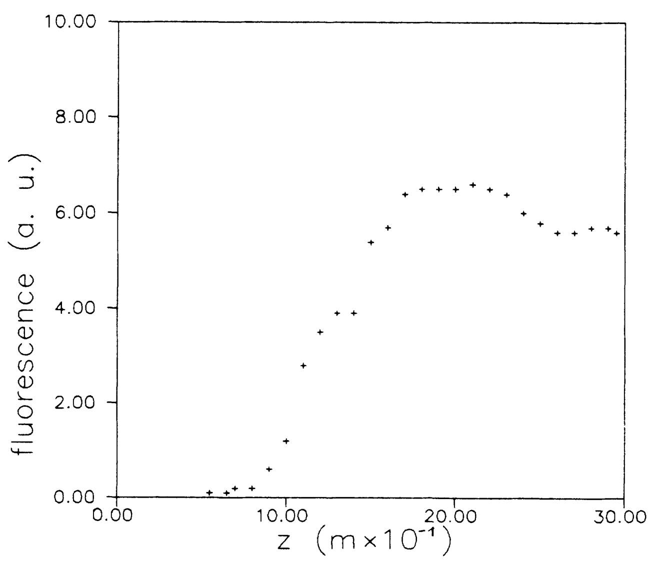

FIG. 13. Cross correlation: the signal S_1 of (10.1) vs z coordinate (along laser).

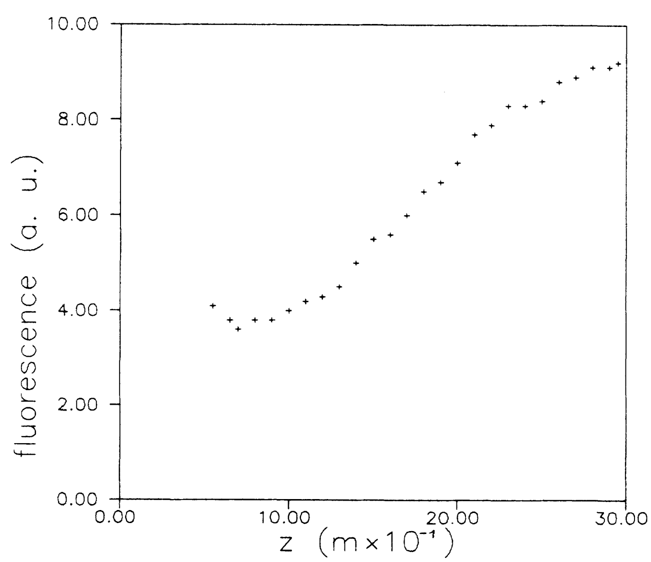

FIG. 15. Signal S_3 of (10.3) vs z coordinate.

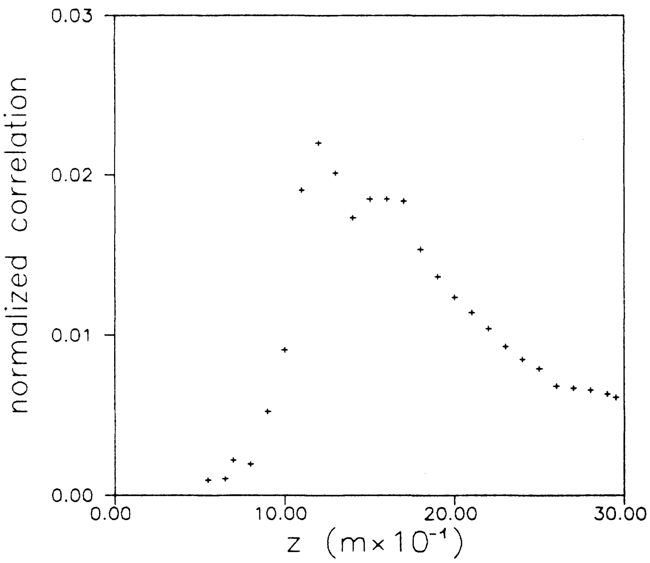

FIG. 16. $d(z)$ of (11), i.e., the signal proportional to the normalized cross correlation; a remarkable growth is seen, although the absolute value is not measured.

As mentioned earlier, the mirror M_0 sufficed for b_2 buildup. The signal S_2 in Fig. 14 shows its intensity along the tube. As a matter of fact, we measured $\rho_c^{(0)} - A_2\langle I_2\rangle$; that is why the signal S_2 drops with the departure from the mirror M_0. The constant $\rho_c^{(0)}$ is unknown but the results turned out to be almost identical for various values of it within a wide range. In Figs. 13–15 our three signals are plotted and the degree of cross correlation (11) is depicted in Fig. 16. It follows from the discussion of Sec. V A that it is to be interpreted as

$$\langle I_1 I_2\rangle + \text{const.} \tag{12}$$

As was mentioned in Sec. II, the sign of the coherent term is difficult to determine if all three levels are pumped. The signal S_1 rises from a low background; it means that the constant of (12) is small compared to $\langle I_1 I_2\rangle$ (if positive at all).

One can see the influence of b_2 on b_1: b_1 also grows towards the propagation direction of b_2. The effect is quite distinct and shows that the role of this kind of pumping b_1 by b_2 is comparable to that of the incoherent one.

If there was not significant correlation between the beams, the signal S_1 would be proportional to the product of the intensities and the plot of Fig. 16 would be flat. Yet, our experimental data form a sharp peak. Hence $\langle I_1 I_2\rangle / (\langle I_1\rangle\langle I_2\rangle)$ is not just a constant along the cavity, but on the contrary, it varies quite distinctly. It proves that the mutual correlation between the two laser fields does exist and evolves in the course of propagation in the active medium, being able to increase remarkably merely by the propagation. The effect is relatively significant, for it manifests itself in the z dependence of simple fluorescence intensity measurements over a distance of typical laser-tube length.

Thus the propagation of beam b_2 results not only in its buildup and an increase of the power of the second (pumped) beam, but also in the buildup of the correlation between the two laser fields. This result also proves that the interaction between the two laser beams cannot be described merely in terms of beam power, but that statistical quantities must be taken into consideration.

The effect we have described is sensitive to the cavity quality Q. For example, moving the common mirror M_0 a few centimeters causes a change in the sharpness of the peak.[12] By comparison of the experimental data and the plots obtained from theory, one can see that they are in good (at least qualitatively) agreement. The quantitative comparison cannot be carried out because of a number of unknown, uncontrollable parameters. The simplicity of our experiment does not allow us to determine and control them. The most important feature of this experiment is the ability to make out the coherent part of the two-photon transitions and to detect the cross correlation of the two photons by a relatively simple spectroscopic technique. Further refinements of the experiment should let us change and control some parameters. Combination of two laser beams should offer a variety of possibilities.

VI. CONCLUSIONS

The treatment of the two-photon laser that we have presented in this paper describes the interaction of two laser beams in an active medium. The laser fields were described by stochastic quantities: average intensities and cross-correlation intensity. We argued the importance of the cross correlation in the phenomena that occur in the laser system. The cross correlation was found to evolve in the course of propagation: the beams can become correlated. The theoretical results were in good qualitative agreement with the experimental results obtained on the He-Ne laser. Both theory and experiment point out that the interaction between laser beams is effectuated not only by their powers but by their mutual correlation matters as well. The phenomena described by the theory are detectable by means of fluorescence measurements. The statistical properties of light change by propagation, which is reflected in fluorescence intensity. Our theory is a phenomenological one and can be considered as a first approximation. Also, the experiment is likely to be the first of a series, because the results seem to be promising.

ACKNOWLEDGMENTS

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APPENDIX

Here we give the values of the coefficients of Eqs. (8) that we used to generate the plots of Figs. 2–11. The second set, given in parentheses, concerns Fig. 10.

First beam: $a_1=2.65$ (2.80), $b_1=20.0$ (20.50), $c_1=24$ (22.90), $d_1^+=0.45$ (0.77), $d_1^-=d_1^+$, $e_1^+=0.15$ (0.17), $e_1^-=0.9e_1^+$.

Second beam: $a_2=2.95$ (3.0), $b_2=3.70$ (3.90), $d_2^+=0.4$ (0.55), $d_2^-=d_2^+$, $e_2^+=0.10$ (0.12), $e_2^-=e_2^+$.

The cross correlation: $\alpha^+=5.20$ (5.40), $\alpha^-=0.23$ (0.18), $\beta^+=3.25$ (3.15), $\beta^-=3.75$ (3.75), $\delta^+=7.60$ (7.65), $\delta^-=4.90$ (4.8), $\gamma^+=23$ (21), $\gamma^-=21$ (17.5), $\epsilon^+=3.0$ (3.0), $\epsilon^-=\epsilon^+$.

Integration intervals were 0–3.12 m (0–3.30 m).

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