Some Consequences of Pump Coherence on Energy Exchange in Nonlinear Optical Processes*

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The finite coherence time of the driving field plays an important role in the rates of energy exchange taking place in nonlinear processes. As a typical example, the problems of the quantum parametric amplifier and frequency converter driven by a fluctuating pump are considered. A closed hierarchy of equations for the moments of the fields is given, which in turn is used for evaluating the average number of photons per mode and the relative variance as functions of time. Significant deviations from the case where the pump field is coherent are found.

The exchange of power between three optical fields in an asymmetric crystal[1] when the "pump" is a random field may deviate significantly from the case in which the coherence time of the driving field is infinite. This occurs, in particular, in the cases of parametric amplification and frequency conversion. These phenomena, which form the basis for a new class of optical devices, have been investigated extensively by assuming as driving pump a well-prescribed harmonic function of time.[2,3] Despite its experimental relevance, no attention has been paid to the fact that the complex amplitude of the laser field, used as a pump, is a centered stochastic process with a finite coherence time which can be considerably shorter than the relevant interaction time. The aim of this Letter is to show that the rate of energy exchange as well as the fluctuations of the two processes mentioned above depend strongly on the coherence time of the pump.

The starting point of our analysis is an equation of motion for the P representation, which has been recently derived.[4,5] Whenever the Hamiltonian of N interacting modes is a polynomial in the creation and annihilation operators whose generic terms are of the form

$$H_{\{m_k,n_k\}}\prod_{k=1}^{N} a_k^{\dagger m_k} a_k^{n_k},$$

we have

$$i\partial P(\{\alpha_k^*,\alpha_k\};t)/\partial t=\Lambda(t)P(\{\alpha_k^*,\alpha_k\};t), \tag{1}$$

where

$$\Lambda(t)=\sum_{\{m_k,n_k\}}H_{\{m_k,n_k\}}\Big[\prod_{k=1}^{N}\exp(|\alpha_k|^2)(-\partial/\partial\alpha_k)^{m_k}\exp(-|\alpha_k|^2)\alpha_k^{n_k} - \prod_{k=1}^{N}\exp(|\alpha_k|^2)(-\partial/\partial\alpha_k^*)^{n_k}\exp(-|\alpha_k|^2)\alpha_k^{*m_k}\Big]. \tag{2}$$

For the parametric amplifier and frequency converter,[2,3] these operators are given in the interaction representation, respectively, by

$$\Lambda(t)=k(t)\left(\frac{\partial^2}{\partial\alpha_1\partial\alpha_2}-\alpha_2^*\frac{\partial}{\partial\alpha_1}-\alpha_1^*\frac{\partial}{\partial\alpha_2}\right)+k^*(t)\left(-\frac{\partial^2}{\partial\alpha_1^*\partial\alpha_2^*}+\alpha_2\frac{\partial}{\partial\alpha_1^*}+\alpha_1\frac{\partial}{\partial\alpha_2^*}\right) \tag{3a}$$

and

$$\Lambda(t)=k(t)\left(\alpha_2^*\frac{\partial}{\partial\alpha_1^*}-\alpha_1\frac{\partial}{\partial\alpha_2}\right)+k^*(t)\left(\alpha_1^*\frac{\partial}{\partial\alpha_2^*}-\alpha_2\frac{\partial}{\partial\alpha_1}\right), \tag{3b}$$

where $k(t)$ is the coupling coefficient induced by the pump between the modes 1 and 2. We assume that $k(t)$ is a centered stationary stochastic process inducing a small change of the P function during its correlation time. In view of this, the Born approximation for the evolution of the $\overline{P}$ [averaged with respect to the $k(t)$ realizations[6]] applies to Eq. (1) which for times t longer than the $k(t)$ correlation time is superceded by

$$\partial\overline{P}(\alpha_1^*,\alpha_2^*,\alpha_1,\alpha_2;t)/\partial t=-\int_0^t\langle\Lambda(t)\Lambda(t-s)\rangle\overline{P}(\alpha_1^*,\alpha_2^*,\alpha_1,\alpha_2;t-s)\,ds. \tag{4}$$

From Eq. (4) it follows that the moments

$$\langle m,n;t\rangle \equiv \int d^2\alpha_1 d^2\alpha_2\, \alpha_1{}^m \alpha_1{}^{*m} \alpha_2{}^n \alpha_2{}^{*n}\, \overline{P}(\alpha_1{}^*, \alpha_2{}^*, \alpha_1, \alpha_2; t) \tag{5}$$

obey the relation

$$d\langle m,n;t\rangle/dt = -\int_0^t ds \int d^2\alpha_1 d^2\alpha_2\, \overline{P}(\alpha_1{}^*, \alpha_2{}^*, \alpha_1, \alpha_2; t)\langle\Lambda(t-s)\Lambda(t)\rangle^\dagger \alpha_1{}^m\alpha_1{}^{*m}\alpha_2{}^n\alpha_2{}^{*n}, \tag{6}$$

where $\langle\Lambda(t-s)\Lambda(t)\rangle^\dagger$ defines an operator which is obtained from $\langle\Lambda(t-s)\Lambda(t)\rangle$ by the substitution $\partial/\partial\alpha_i \to -\partial/\partial\alpha_i$. Because of the structure of our Hamiltonians, Eq. (6) defines a closed hierarchy of integro-differential equations,

$$d\langle m,n;t\rangle/dt = \int_0^t ds\, g(s)[m^2n^2\langle m-1,n-1;t-s\rangle + m^2(2n+1)\langle m-1,n;t-s\rangle + n^2(2m+1)\langle m,n-1;t-s\rangle + m^2\langle m-1,n+1;t-s\rangle + n^2\langle m+1,n-1;t-s\rangle + (2mn+m+n)\langle m,n;t-s\rangle], \tag{7a}$$

for the parametric amplification, and

$$d\langle m,n;t\rangle/dt = \int_0^t ds\, g(s)[m^2\langle m-1,n+1;t-s\rangle + n^2\langle m+1,n-1;t-s\rangle - (2mn+m+n)\langle m,n;t-s\rangle] \tag{7b}$$

for the frequency conversion, $g(s)$ standing for $2\langle k(t)k^*(t-s)\rangle$. This system of equations can be solved by applying the Laplace-transform technique whenever the spectral profile of the pump is known. In the following we shall consider a Lorentzian laser profile $[g(s) = 2k_0{}^2e^{-\gamma s}]$, and we shall give the expression for the first two moments. We note that in practical situations we can have $\eta \equiv k_0/\gamma \ll 1$ (i.e., a small fractional change during a "coherence interval" γ^{-1}). To the lowest significant order in η we have for the parametric amplifier (assuming $\langle n_{20}\rangle = 0$)

$$\langle n_1(t)\rangle = \tfrac{1}{2}(\langle n_{10}\rangle + 1)\exp(4\eta k_0 t) + \tfrac{1}{2}\langle n_{10}\rangle - \tfrac{1}{2}, \tag{8a}$$

$$\langle n_1{}^2(t)\rangle = \tfrac{1}{6}(2\langle n_{10}{}^2\rangle - 5\langle n_{10}\rangle + 5) + \tfrac{1}{2}(\langle n_{10}{}^2\rangle - 3)\exp(4\eta k_0 t) + \tfrac{1}{6}(\langle n_{10}{}^2\rangle + 5\langle n_{10}\rangle + 2)\exp(12\eta k_0 t), \tag{8b}$$

and for the frequency converter (assuming $\langle n_{10}\rangle = 0$)

$$\langle n_1(t)\rangle = \tfrac{1}{2}\langle n_{20}\rangle[1 - \exp(4\eta k_0 t)], \tag{9a}$$

$$\langle n_1{}^2(t)\rangle = \tfrac{1}{6}(2\langle n_{20}{}^2\rangle - \langle n_{20}\rangle) - \tfrac{1}{2}\langle n_{20}{}^2\rangle\exp(-4\eta k_0 t) + \tfrac{1}{6}(\langle n_{20}{}^2\rangle + \langle n_{20}\rangle)\exp(-12\eta k_0 t), \tag{9b}$$

having introduced $\langle n(t)\rangle = \langle a^\dagger a\rangle$ and $\langle n^2(t)\rangle = \langle a^\dagger a a^\dagger a\rangle$. Equations (8) and (9) can be compared with the corresponding ones for the coherent pump ($\eta \gg 1$),[2] which for $\langle n(t)\rangle$ read

$$\langle n_1(t)\rangle = \langle n_{10}\rangle\cosh^2 k_0 t + \sinh^2 k_0 t \tag{10a}$$

and

$$\langle n_1(t)\rangle = \langle n_{20}\rangle\sin^2 k_0 t. \tag{10b}$$

A general consequence of the pump randomness is a reduction of the growth rate by a factor $\eta \ll 1$ so that all the processes are significantly slowed down. For what concerns the frequency converter, it is interesting to note that only one half of the power in the signal can be converted asymptotically into the idler, in accordance with the statistical equipartition principle. In the "coherent-pump" case complete power exchange between the idler and signal fields occurs periodically.

With the use of Eqs. (8) and (9) the normalized variance $v = (\langle n^2\rangle - \langle n\rangle^2)/\langle n\rangle^2$ can be calculated. Since time intervals of physical interest often satisfy the condition $\eta k_0 t \ll 1$, the exponential occurring in Eqs. (8) and (9) can be approximated by linear functions of time, thus giving for the frequency converter (in the limit $\langle n_{20}\rangle \gg 1$)

$$v = \frac{\langle n_{20}{}^2\rangle - \langle n_{20}\rangle^2}{\langle n_{20}\rangle^2} + \frac{\langle n_{20}{}^2\rangle}{\langle n_{20}\rangle^2}. \tag{11}$$

Equation (11) shows that v changes from 0 to 1 if the signal is initially coherent $(\langle n_{20}{}^2\rangle = \langle n_{20}\rangle + \langle n_{20}\rangle^2)$ or from 1 to 3 if the signal is initially Gaussian $(\langle n_{20}{}^2\rangle = \langle n_{20}\rangle + 2\langle n_{20}\rangle^2)$. In the same limit $(\langle n_{10}\rangle \gg 1)$ the v for the parametric amplifier can be shown not to change appreciably during the time interval of interest.

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Study of Heat Diffusion in Transparent Media by Means of a Thermal-Lens Effect

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The thermal-lens effect is used to study the dynamics of heat diffusion in transparent media. Values of the thermal diffusivity and thermal conductivity are obtained for several liquids.

A beam of light passing through a transparent medium modifies its index of refraction. Four different mechanisms are mainly responsible for this: (a) the Kerr effect, related to molecular reorientation; (b) electrostriction; (c) the electrocaloric effect, which is a thermodynamical consequence of the existence of a nonvanishing $d\epsilon/dT$ term; and (d) nonradiative absorption, which creates heating all along the path of the light beam in absorbing dielectrics.

The relative importance of these mechanisms depends on both the nature of the dielectric and the time scale considered. For instance, at times of the order of 10^{-8} sec or less, only the Kerr effect, which does not involve density modification, needs be considered. On the other hand for a dielectric having an absorption coefficient α of the order of 10^{-4} to 10^{-3} cm^{-1} and for times $\gtrsim 10^{-1}$ sec, the nonradiative absorption gives an effect 10^4 to 10^8 times greater than the three other mechanisms. We conclude that for the *times considered in the following experiments*, which are typically from 1 to 20 sec, only this last mechanism needs to be considered.

As shown by Gordon *et al.*,[1] the nonradiative absorption gives rise to the so-called thermal-lens effect. Indeed the laser beam yields localized heating along its path and a consequent transverse gradient of the index of refraction n, which is usually minimum at the center of the beam, thus creating a diverging lens in the dielectric. Leite, Morre, and Whinnery[2] have used this effect for the measurements of very low absorbancies. We try to use it to study the dynamics of heat diffusion in transparent media and eventually measure their absorbancies.

Theoretical background and experimental setup.—As shown in Ref. 1, when an infinite medium is illuminated at time $t=0$ along the z axis with a light beam of Gaussian intensity $P=P_0(2/\pi\omega_0^2)\times\exp(-2r^2/\omega_0^2)$, in the limit of small absorbencies and local heating, the temperature distribution is given in terms of exponential integrals by

$$\delta T(r,z,t)=\frac{\alpha P_0}{4\pi\Lambda}\left[\mathrm{Ei}\left(-\frac{2r^2}{\omega_0^2}\right)-\mathrm{Ei}\left(-\frac{2r^2}{\omega_0^2+8Dt}\right)\right]e^{-\alpha z},$$

where α is the absorption coefficient, Λ the thermal conductivity, and $D=\Lambda\rho C_p$ the coefficient of thermal diffusion, with C_p the specific heat and ρ the density.

This temperature distribution gives a corresponding distribution of refractive index,

$$\delta n(r,z,t)=\frac{dn}{dT}\delta T(r,z,t)=\frac{\alpha P_0}{4\pi\Lambda}\frac{dn}{dT}\left[\mathrm{Ei}\left(-\frac{2r^2}{\omega_0^2}\right)-\mathrm{Ei}\left(-\frac{2r^2}{8D(t+t_c)}\right)\right]e^{-\alpha z},$$

where $t_c=\omega_0^2/8D$ is a characteristic time.

As stressed in Ref. 1, this model has the weakness that $\delta T\to\infty$ when $t\to\infty$. This is because the heat dissipation on the cell boundaries has not been taken into account. However we may expect that the