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Time Dependence of Wave-Mixing Processes under Pulsed Resonant Excitation in Three-Level Systems

By

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A theoretical analysis is given of wave-mixing processes in three-level systems excited by nanosecond pulses close to a two-photon resonance. It is shown that the lifetime of the incoherent quasiparticle population influences the time dependent generation of the coherent signal.

Es wird eine theoretische Analyse der Wellenmisch-Prozesse in Drei-Niveau-Systemen angegeben, die durch Nanosekundenimpulse in der Nähe einer Zwei-Photonenresonanz angeregt werden. Es wird gezeigt, daß die Lebensdauer der inkohärenten Quasiteilchenbesetzung die zeitabhängige Generation des kohärenten Signals beeinflußt.

1. Introduction

In a foregoing article [1], we have discussed theoretically the competition between coherent and incoherent scattering processes in degenerate signal generation. We shall study now the consequences of this competition for a real system [2], in which the transient nonlinear polarization can be calculated explicitly from the transition rates between the different states [3]. We consider here the case of CuCl, for which experimental results have been obtained recently.

Time-resolved experiments of signal generation have been mainly performed under picosecond excitation condition [4 to 6], in which dephasing times T_2 of the resonantly generated states can be studied. Under nanosecond excitation conditions [2], structures in the temporal shape of the signal appear which have different origins. First, the complex nonlinear dielectric function does not follow instantaneously the shape of the exciting pulses but shows a memory effect due to the population lifetime T_1. Secondly, the generation rate varies between oscillatory attenuation and gain due to a competition between coherent and incoherent scattering processes.

2. Time-Dependent Signal Generation

The nonlinear wave equation for the i-th component of an electric field $E_i(z, t)$ reads [7]

$$\frac{\partial^2 E_i(z, t)}{\partial z^2} - \mu_0 \sigma(\omega) \frac{\partial E_i(z, t)}{\partial t} - \mu_0 \varepsilon_l(\omega) \frac{\partial^2 E_i(z, t)}{\partial t^2} = \mu_0 \frac{\partial^2}{\partial t^2} P^i_{nl}(z, t) , \tag{1}$$

where $\sigma(\omega)$ is the conductivity of the system, $\varepsilon_l(\omega) = \varepsilon_0\big(1 + \chi_l(\omega)\big)$ the linear dielectric function, μ_0 and ε_0 denote the permeability and permittivity constants, respec-

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tively, and $P^i_{\mathrm{nl}}(z, t)$ the nonlinear polarization. Let us consider now only three frequency degenerated fields ($i \in$ p, s, t), denoting the pump, signal, and test beams, respectively, propagating almost parallely to the z-direction, and let us neglect transverse field effects. The field components may then be described by plane waves

$$E_i(z, t) = \tfrac{1}{2} \left(E^0_i(z, t)\, \mathrm{e}^{i(\omega t - k_i z)} + \text{c.c.}\right) , \tag{2}$$

where c.c. denotes complex conjugation. The field amplitudes are assumed to vary slowly in time and space when compared to the frequency ω and the wave vector magnitude k_i. Since we restrict ourselves to fields with the same frequency ω, we consider only the nonlinear polarization component $P^{i,\omega}_{\mathrm{nl}}(z, t)$ (i.e. we neglect all contributions which give rise to signals of other frequencies). $P^{i,\omega}_{\mathrm{nl}}(z, t)$ is given by the following expression [1]:

$$P^{i,\omega}_{\mathrm{nl}}(z, t) = \tfrac{1}{2} \left[\sum_j \chi^{ij(0)}_{\mathrm{nl}}(z, t, \omega)\, E^0_j\, \mathrm{e}^{i(\omega t - k_j z)} + \chi^{ij(2)*}_{\mathrm{nl}}(z, t, \omega)\, E^{0*}_j\, \mathrm{e}^{i[\omega t - (2k_p - k_j) z]}\right] . \tag{3}$$

Equation (3) is obtained from the definition of the nonlinear polarization as a convolution product of the nonlinear susceptibility $\chi^{ij}_{\mathrm{nl}}(z, t, t')$ and of the electric field $E_j(z, t')$. If we consider local spatial response for simplicity, we may write

$$P^i_{\mathrm{nl}}(z, t) = \sum_j \int\limits_{-\infty}^{t} \chi^{ij}_{\mathrm{nl}}(z, t, t')\, E_j(z, t')\, \mathrm{d}t' . \tag{4}$$

Introducing $\tau = t - t'$ as a new variable and separating $\chi^{ij}_{\mathrm{nl}}(z, t, \tau)$ into slow and fast varying terms, we obtain

$$\chi^{ij}_{\mathrm{nl}}(z, t, \tau) = \sum_n \chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)\, \mathrm{e}^{in(\omega t - kz)} , \tag{5}$$

and with (2) and (4),

$$\begin{aligned} P^i_{\mathrm{nl}}(z, t) = \tfrac{1}{2} \sum_{jn} \int\limits_0^{\infty} \chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)\, \{E^0_j(z, t - \tau)\, \mathrm{e}^{i[(n+1)\omega t - (nk + k_j) z]} \times \\ \times \mathrm{e}^{-i\omega\tau} + E^{0*}_j(z, t - \tau)\, \mathrm{e}^{i[(n-1)\omega t - (nk - k_j) z]}\, \mathrm{e}^{+i\omega\tau}\}\, \mathrm{d}\tau . \end{aligned} \tag{6}$$

If $E^0_j(z, t - \tau)$ is slowly varying on a time-scale where $\chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)$ is strongly peaked [3], the field amplitudes can be considered as constant in (6) and $P^i_{\mathrm{nl}}(z, t)$ can be approximated to

$$\begin{aligned} P^i_{\mathrm{nl}}(z, t) = \tfrac{1}{2} \Big[\sum_{jn} \Big[E^0_j(z, t)\, \mathrm{e}^{i[(n+1)\omega t - (nk + k_j) z]} \int\limits_0^{\infty} \chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)\, \mathrm{e}^{-i\omega\tau}\, \mathrm{d}\tau + \\ + E^0_j(z, t)^*\, \mathrm{e}^{i[(n-1)\omega t - (nk - k_j) z]} \int\limits_0^{\infty} \chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)\, \mathrm{e}^{+i\omega\tau}\, \mathrm{d}\tau\Big] . \end{aligned} \tag{7}$$

We may now define a time and frequency dependent susceptibility component by

$$\chi^{ij(n)}_{\mathrm{nl}}(z, t, \omega) = \int\limits_0^{\infty} \chi^{ij(n)}_{\mathrm{nl}}(z, t, \tau)\, \mathrm{e}^{-i\omega\tau}\, \mathrm{d}\tau . \tag{8}$$

When using the relation $\chi(z, t, \omega) = \chi^*(z, t, -\omega)$, we obtain for $P^i_{\mathrm{nl}}(z, t)$

$$\begin{aligned} P^i_{\mathrm{nl}}(z, t) = \tfrac{1}{2} \Big[\sum_{jn} \chi^{ij(n)}_{\mathrm{nl}}(z, t, \omega)\, E^0_j(z, t)\, \mathrm{e}^{i[(n+1)\omega t - (nk + k_j) z]} + \\ + \chi^{ij(n)*}_{\mathrm{nl}}(z, t, \omega)\, E^{0*}_j(z, t)\, \mathrm{e}^{i[(n-1)\omega t - (nk - k_j) z]}\Big] . \end{aligned} \tag{9}$$

When keeping in (9) only the terms oscillating with the frequency ω, we obtain (3). We are now in the position to solve (1) in the slowly varying envelope approximation. By doing this, we neglect all time derivatives of the slowly varying functions $E_j^0(t)$ and $\chi_{\mathrm{nl}}^{ij(n)}$ with respect to ω. In addition, we assume that $(\partial^2/\partial z^2)\, E_j^0 \ll k_j(\partial/\partial z)\, E_j^0$. When using the dispersion relation $\mu_0\varepsilon_1\omega^2 = k_i^2$ and restricting to the three fields (p, s, t) discussed above, we obtain

$$-2ik_i \frac{\partial}{\partial z} E_i^0(z, t) - i\omega\mu_0\sigma E_i^0(z, t)\, \mathrm{e}^{i(\omega t - k_i z)} + \text{c.c.} =$$

$$= -\frac{1}{2}\, \mu_0\omega^2 \left\{\sum_j \chi_{\mathrm{nl}}^{ij(0)}(z, t, \omega)\, E_j^0(z, t)\, \mathrm{e}^{i(\omega t - k_j z)} + \right.$$

$$\left. + \sum_j \chi_{\mathrm{nl}}^{ij(2)*}(z, t, \omega)\, E_j^0(z, t)^*\, \mathrm{e}^{i[\omega t - (2k - k_j) z]}\right\} + \text{c.c.}, \tag{10}$$

where we still have to respect the phase-matching condition

$$\Delta k = 0 = 2k - k_j - k_i \tag{11}$$

when summing over the fields with index j.

Let us now assume that the test and the signal fields are so weak that they do not introduce changes in the nonlinear susceptibility [8]. Therefore, pump, signal, and test beams are subject to the same nonlinearities, generated by the pump beam. With this approximation, we identify k with k_{p} and obtain for the pump beam

$$\frac{\partial}{\partial z} E_{\mathrm{p}}^0(z, t) = -\frac{\mu_0\omega\sigma}{2k} E_{\mathrm{p}}^0(z, t) + \chi_{\mathrm{nl}}^{\mathrm{pp}(0)}(z, t, \omega)\, \frac{\mu_0\omega^2}{2ik_{\mathrm{p}}} E_{\mathrm{p}}^0(z, t) +$$

$$+ \chi_{\mathrm{nl}}^{\mathrm{pp}(2)*}(z, t, \omega)\, \frac{\mu_0\omega^2}{2ik_{\mathrm{p}}} E_{\mathrm{p}}^{0*}(z, t)\,. \tag{12}$$

It follows for the test beam:

$$\frac{\partial}{\partial z} E_{\mathrm{t}}^0(z, t) = -\frac{\mu_0\omega\sigma}{2k_{\mathrm{t}}} E_{\mathrm{t}}^0 + \chi_{\mathrm{nl}}^{\mathrm{tt}(0)}(z, t, \omega)\, \frac{\mu_0\omega^2}{2ik_{\mathrm{t}}} E_{\mathrm{t}}^0 + \chi_{\mathrm{nl}}^{\mathrm{ts}(2)*}(z, t, \omega)\, \frac{\mu_0\omega^2}{2ik_{\mathrm{t}}} E_{\mathrm{s}}^{0*}\,. \tag{13}$$

Since the phase-matching condition is almost fulfilled in (10), the test and signal beams are emitted symmetrically with respect to the pump beam. Then, the equation describing the signal field is obtained by simply interchanging the indices t and s in (13).

As discussed in more detail in 1, the first term on the right-hand side of (12) and (13) gives rise to the linear absorption, the second one (proportional to $\chi_{\mathrm{nl}}^{ii(0)}$) to the nonlinear absorption and dispersion, induced by the pump beam. In these terms, the excited states involved in the process have not necessarily to be coherent with the interacting beams. The last term, on the contrary, represents a parametric process in which the pump and the test beams generate the signal beams coherently. Then, only coherent states contribute to this generation (via $\chi_{\mathrm{nl}}^{\mathrm{st}(2)}(z, t)$). Therefore, in order to solve (12) and (13), we have to know the coherent and incoherent contributions of $\chi_{\mathrm{nl}}^{ii(0)}$ and the coherent one of $\chi_{\mathrm{nl}}^{\mathrm{st}(2)}$ separately.

3. Coherent and Incoherent Nonlinear Susceptibilities in CuCl

In order to determine the coherent and incoherent parts of the nonlinear susceptibilities, we consider a three-level system which is well adapted to describe CuCl. As discussed in more detail in [9, 10], it consists of the crystal ground state $|1\rangle$, the exciton state $|2\rangle$, and the biexciton state $|3\rangle$. The transitions between states $|1\rangle$ and $|2\rangle$ and $|2\rangle$ and $|3\rangle$ are dipole allowed for linear polarizations, those between $|1\rangle$ and $|3\rangle$ are allowed for two-photon transitions. The transition elements are $\mu_{ex} = \langle 2| \mu |1\rangle$ and $\mu_{bi} = \langle 3| \mu |2\rangle$, respectively, μ being the dipole operator. In the framework of the density matrix formalism, the polarization $P(t)$ can be expressed by

$$P(t) = N \operatorname{tr} [\varrho(t) \mu] , \tag{14}$$

ϱ_{ij} being the elements of the density matrix and N the density of unit cells in the crystal. We may calculate the time dependence of ϱ from the Schrödinger equation using

$$\frac{\partial}{\partial t} \varrho_{ij}(t) = \frac{i}{\hbar} [\varrho, H]_{ij} - \varrho_{ij} \Gamma_{ij} , \tag{15}$$

where H is the Hamiltonian of the system and Γ_{ij} denotes the different damping constants. Let us discuss the meaning of the dampings. In a first stage, we consider the quasi-particle populations. In order to calculate the expectation value of the dipole operator, we have to build the ensemble average over these populations. Γ_{ii} then represents the energy transfer from the exciton–biexciton system towards a heat bath and $T_i = 1/\Gamma_{ii}$ is the corresponding decay time due to inelastic collisions. These dampings do not give information on the time evolution of the actual states which are excited coherently. In order to know their time evolution, we have to define a different ensemble average made only of equivalent states. We denote their decay time by $\tilde{T}_i = 1/\tilde{\Gamma}_{ii}$. It involves both elastic and inelastic collision processes. In a two-level system, these lifetimes of quasi-stationary, coherent states are related to the phase (transverse) relaxation time T_2 via: $1/T_2 = 1/2(\tilde{T}_{11} + 1/\tilde{T}_{22})$. Here, we adopt the general definition: $\Gamma_{ij} = 1/2(1/\tilde{T}_{ii} + 1/\tilde{T}_{jj})$ for $i \neq j$.

We now separate ϱ_{ij} in slowly and rapidly varying contributions by

$$\varrho_{ij}(t, z) = \sum_n \varrho_{ij}^n(z, t) \, e^{in(\omega t - kz)} \tag{16}$$

and express (14) in the form

$$P(z, t) = N \sum_n [(\varrho_{12}^n(z, t) + \varrho_{21}^n(z, t)) \mu_{ex} + (\varrho_{23}^n(z, t) + \varrho_{32}^n(z, t)) \mu_{bi}] \, e^{in(\omega t - kz)} . \tag{17}$$

We now compare (17) to (3) and retain again only terms that oscillate with frequency ω. We then obtain (when using the same approximations as those leading to (12) and (13)) for the i-th field component,

$$\begin{aligned} &N[(\varrho_{12}^1(z, t) + \varrho_{21}^1(z, t)) \mu_{ex} + (\varrho_{23}^1(z, t) + \varrho_{32}^1(z, t)) \mu_{bi}] = \\ &= \chi_l E_i^0(z, t) + \chi_{nl}^{ii(0)}(z, t) \, E_i^0(z, t) + \chi_{nl}^{ij(2)*}(z, t) \, E_j^{0*}(z, t) , \end{aligned} \tag{18}$$

where χ_l denotes the linear part of the susceptibility. As stated above, $\chi_{nl}^{ij(2)*}$ contains only coherent contributions while all the other terms do not distinguish between coherent and noncoherent parts. In order to separate both contributions, we may first calculate the coherent part of the susceptibility. To do this, we build the ensemble average made of equivalent states and consider in (15) Γ_{ij} ($i \neq j$) and Γ_{ii}.

Equations (15) and (18) then lead to a system of coupled differential equations in time, which is solved numerically as discussed in 3. We thus obtain the coherent part of the susceptibility. It is important to notice that

$$E_i^0(z, t) \, \chi_{nl}^{ii(0)}|_{coh} = E_i^0(z, t) \, \chi_{nl}^{ii(2)}(z, t) \tag{19}$$

(as it is assumed usually when treating degenerate signal generation [7, 8, 11]). In order to calculate the actual polarization of the system, we have to consider population effects. Therefore, we solve in a second step (15) and (17), taking again Γ_{ij} $(i \neq j)$ as before, but for diagonal elements the population dampings Γ_{ii}.

We thus determine from (18) and (19) the nonlinear susceptibility part $\chi_{\mathrm{nl}}^{ii(0)}(z, t)$. As discussed in [3], the latter term shows on a nanosecond time scale the memory effect due to the lifetimes of the populations. $\chi_{\mathrm{nl}}^{ij(2)}$, on the contrary, follows the pulse excitation on this time scale.

4. Temporal Pulse Shape of Signal Emission

In order to discuss the pulse shape of the signal emission, let us adopt a mean field approximation here. We assume the incident pump pulse intensity $I_{\mathrm{p}}^0(t)$ to have Gaussian shape of 4 ns halfwidth (FWHM). If $\alpha(t)$ is the nonlinear absorption resulting from (12), we assume the pump beam intensity inside the crystal to have the mean value (independent of z)

$$\bar{I}_{\mathrm{p}}(t) = \frac{I_{\mathrm{p}}^0(t)}{l\alpha(t)} \left(1 - \mathrm{e}^{-\alpha(t)l}\right), \tag{20}$$

l being the sample thickness, $\alpha(t)$ and $I_{\mathrm{p}}(t)$ are determined self-consistently [12] from (15), (18), and (20). In this approximation, $\chi_{\mathrm{nl}}^{ii(0)}(t)$ and $\chi_{\mathrm{nl}}^{ij(2)}(t)$ are no longer functions of z since they are determined by the pump beam intensity only. Equation (13) gives rise to a system of coupled differential equations for the propagating pump and test beams, which have the form

$$\frac{\mathrm{d}x'}{\mathrm{d}z} = a'x' - a''x'' + b'y' + b''y'', \qquad \frac{\mathrm{d}x''}{\mathrm{d}z} = a''x' + a'x'' + b''y' - b'y'',$$
$$\frac{\mathrm{d}y'}{\mathrm{d}z} = b'x' + b''x'' + a'y' - a''y'', \qquad \frac{\mathrm{d}y''}{\mathrm{d}z} = b''x' - b'x'' + a''y' + a'y'' \tag{21}$$

with $E_{\mathrm{t}} = x' + ix''$, $E_{\mathrm{s}} = y' + iy''$, and

$$a' = -\frac{\omega\mu\sigma}{2k} + \frac{\mu_0\omega^2}{2K} \operatorname{Im} \chi_{\mathrm{nl}}^{ii(0)}(t), \qquad a'' = -\frac{\mu_0\omega^2}{2k} \operatorname{Re} \chi_{\mathrm{nl}}^{ii(0)}(t),$$
$$b' = -\frac{\mu_0\omega^2}{2k} \operatorname{Im} \chi_{\mathrm{nl}}^{ij(2)}(t), \qquad b'' = -\frac{\mu_0\omega^2}{2k} \operatorname{Re} \chi_{\mathrm{nl}}^{ij(2)}(t), \tag{22}$$

$k \approx k_{\mathrm{p}} \approx k_{\mathrm{t}} \approx k_{\mathrm{s}}$ is the wave vector magnitude of the fields.

Considering the initial conditions at $z = 0$ which are $x'(0) = E_{\mathrm{t}}^0(0, t)$, $x''(0) = y'(0) = y''(0) = 0$, we obtain a solution for the intensities of the beams which is a function of z,

$$I_{\mathrm{s}} = |y'(z)|^2 + |y''(z)|^2 = E_{\mathrm{t}}^2\, \mathrm{e}^{+2a'z}\, \frac{b'^2 + b''^2}{|\lambda|^2} \sinh^2(|\lambda|\, z) \tag{23}$$

and

$$I_{\mathrm{t}} = E_{\mathrm{t}}^2\, \mathrm{e}^{+2a'z} \left(\cosh^2(|\lambda|\, z) + \left(\frac{a''}{|\lambda|}\right)^2 \sinh^2(|\lambda|\, z)\right) \tag{24}$$

for $\lambda^2 = -a''^2 + b''^2 + b'^2 > 0$.

In the case $\lambda^2 < 0$, one has to replace $(\sinh(|\lambda|\, z))$, $\cosh(|\lambda|\, z)$ by $(\sin(|\lambda|\, z)$ and $\cos(|\lambda|\, z))$, respectively.

As we have discussed in [1], this result shows that we may obtain gain or attenuated oscillation for the signal and test beam intensities, depending on the value of λ^2. This quantity is constant under stationary excitation conditions. In order to demonstrate the characteristic features of (23) and (24), we have calculated I_s and I_t (for a fixed value of $b'^2 + b''^2 = 10^8\ \text{cm}^{-2}$ and $a' = 10^3\ \text{cm}^{-1}$ and for different values of a'' as functions of z in the region $0 < z < 10\ \mu\text{m}$. The result is shown in Fig. 1 a and b, for $I_s(z)$ and in Fig. 2 a, b for $I_t(z)$ in the oscillation region (Fig. 1a and 2a for $\lambda^2 < < 0$) and in the gain region (Fig. 1b and 2b for $\lambda^2 > 0$). (Note the different scales on the ordinates.) It is interesting to notice that not only the signal and the transmitted test beam amplitudes decrease with increasing values of a'' but that also the period of the oscillations changes with a''. Since a'' is intensity dependent, the signal intensity may even diminish with increasing intensity for a fixed value of z.

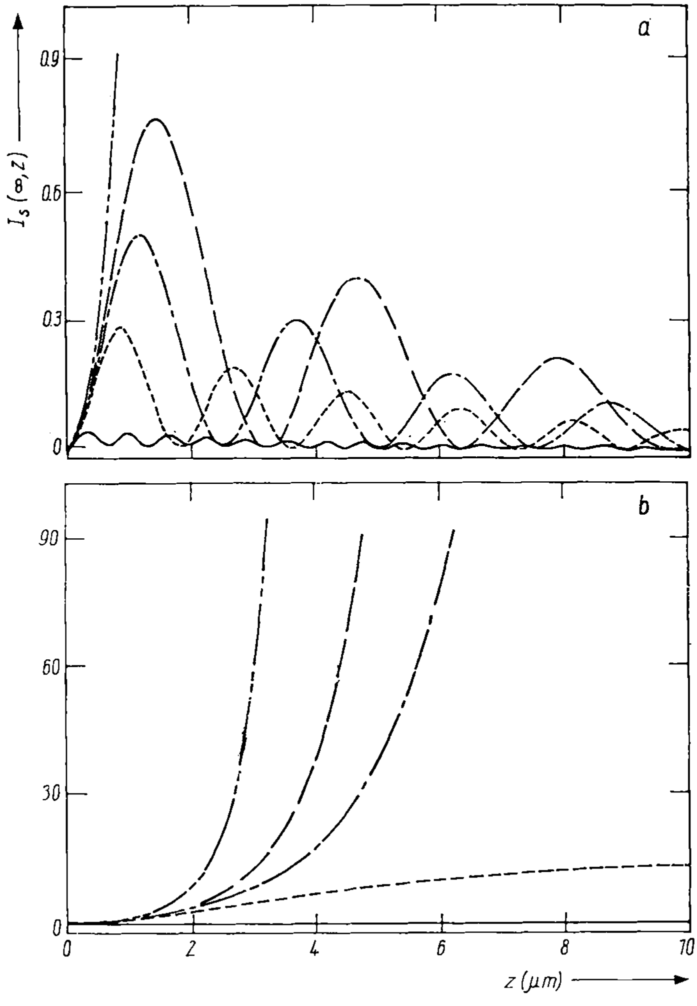

Fig. 1. Stationary signal intensity $I_s(\infty, z)$ for different values of a'' and a fixed value of $(b'^2 + b''^2) = 10^8\ \text{cm}^{-2}$ in a) the oscillatory regime ($\lambda^2 < 0$) and b) the gain region ($\lambda^2 > 0$); a) $a'' = 10^3$ (—··—), 1.4×10^4 (— — —), 1.6×10^4 (—·—) 2×10^4 (––––), $5 \times 10^4\ \text{cm}^{-1}$ (———); b) $a'' = 10^3$ (—··—), 8×10^3 (— — —), 9×10^3 (—·—), 10^4 (––––), $5 \times 10^4\ \text{cm}^{-1}$ (———)

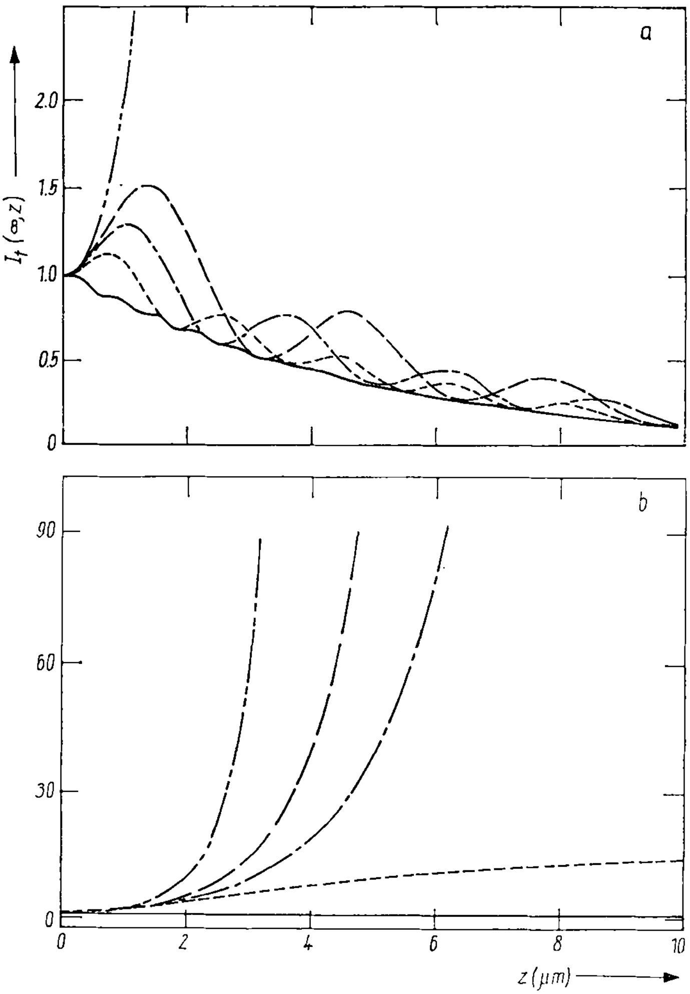

Fig. 2. As Fig. 1, but for the test beam $I_t(\infty, z)$

Concerning pulsed excitation, the situation is even more complicated since a' and a'' show a memory effect, while b' and b'' follow the exciting pulse shape on a nanosecond time scale [3]. Therefore, λ^2 depends strongly on time and may vary during the pulse from $\lambda^2 > 0$ to $\lambda^2 \leqq 0$. To demonstrate this, we have calculated a', a'', b', and b'' for a Gaussian pulse of 4 ns duration and 6 MW/cm^2 maximum intensity, using the three-level model developed for CuCl [3]. Fig. 3 shows the results, using $\hbar\Gamma_{ij} = 10^{-4}$ eV ($i \neq j \in (1, 2, 3)$), $\hbar\Gamma_{ii} = 10^{-6}$ eV, and $\hbar\tilde{\Gamma}_{ii} = 10^{-4}$ eV. These values correspond to dephasing times T_2 of about 6 ps and radiative lifetimes T_1 of about 600 ps. These values have been determined experimentally at low intensities of excitation [13]. The system is excited close to the biexciton resonance at $\hbar\omega = 3.1857$ eV. Fig. 4a shows the resulting pulse shapes of the signal beam and Fig. 4b the generation rate $I_s(t, l)/I_t(t, 0)$. Similarly, Fig. 5a gives that of the transmitted test beam I_t and $I_t(t, l)/I_p(t, 0)$, assuming a crystal length of $l = 10\,\mu$m. When varying $2\hbar\omega$ through the biexciton resonance, the temporal shapes of the transmitted and signal

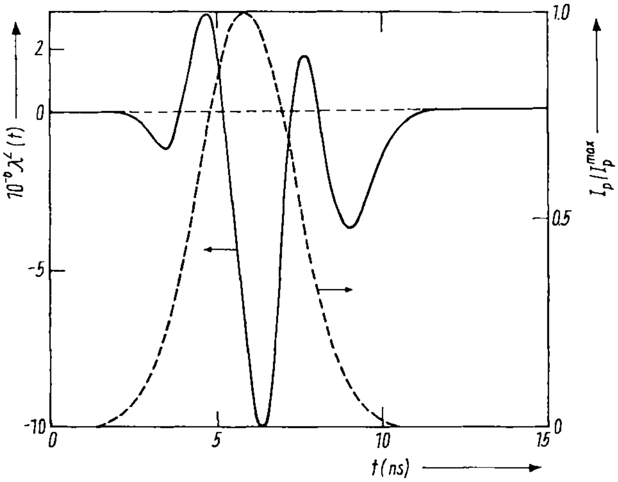

Fig. 3. λ^2 (full line) as function of time under excitation with a Gaussian pulse of 4 ns duration (FWHM) (dashed line)

pulses vary a lot as it was found experimentally. In addition, the integrated signal intensity (excitation spectrum) changes. This is shown in Fig. 6 for a maximum pump intensity $I_p^{max} = 6$ MW/cm^2, assuming a purely coherent susceptibility ($\hbar\Gamma_{ii} = \hbar\tilde{\Gamma}_{ii} = 10^{-4}$ eV, dashed line).

If incoherent processes become important (full line in Fig. 6, calculated using $\hbar\Gamma_{ii} = 10^{-5}$ eV, $\hbar\tilde{\Gamma}_{ii} = \hbar\Gamma_{ij} = 10^{-4}$ eV), the excitation spectrum becomes strongly asymmetric. In any case, the minimum at $\hbar\omega_p = 3.186$ eV, i.e. at half the biexciton energy, is due to the strong absorption of the pump beam and the reabsorption of the test and the signal beams. This effect leads, at the same time, to an increase of the incoherent part of the susceptibility over the coherent one. The asymmetry in the

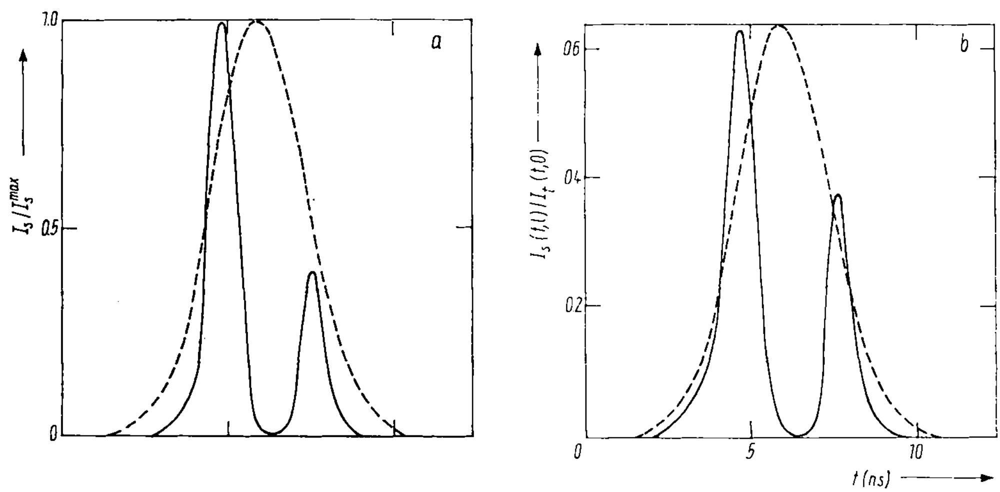

Fig. 4. a) Signal emission $I_s(t, l)$ (full line) corresponding to the conditions of Fig. 3 and b) generation rate $I_s(t, l)/I_t(t, 0)$. The incident pulse shape is indicated by the dashed line

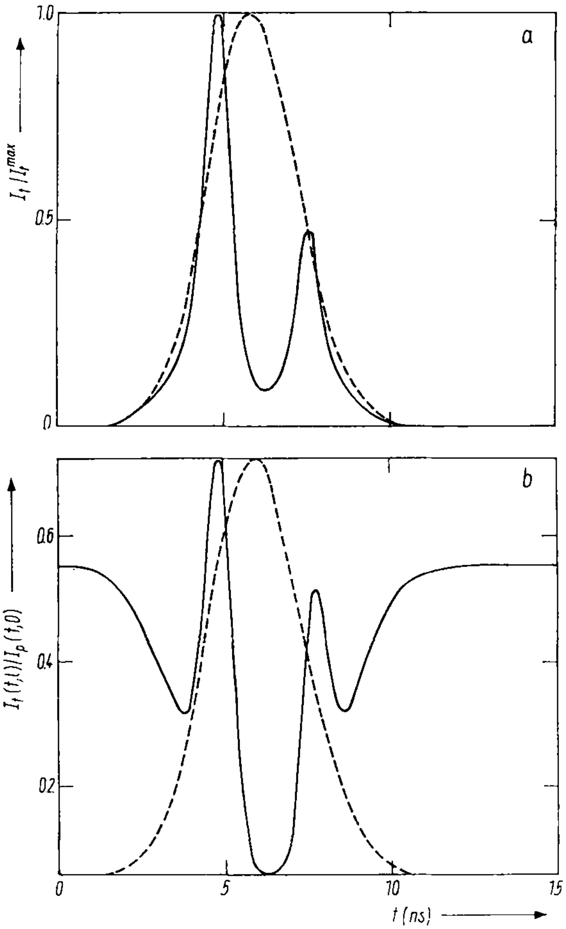

Fig. 5. As Fig. 4, but for $I_t(t, l)$

excitation spectrum, ascribed previously to a Fano-type effect of biexciton decay [11], is here attributed to the asymmetry of a'' with respect to the biexciton resonance.

5. Conclusion

We have shown that coherent and incoherent processes give rise to different terms in the nonlinear susceptibility which compete with each other on a nanosecond time scale. The competition shows up in the generation of a signal beam and in the amplification of a test beam due to a pump beam. It leads to important temporal structures in the pulse shapes. This memory effect is due to the finite lifetime of the quasi-particles, created during the excitation. From this discussion, it is evident that the actual shape of the generated signal depends on several parameters: the frequency and intensity of excitation, the sample thickness, and the intrinsic sample properties as dispersion and absorption.

We explain the asymmetry of the excitation spectrum, which has been observed experimentally, as being due to the competition between coherent and noncoherent parts of the nonlinear susceptibility, and the minimum at $E_{bi}/2$ as being due to a strong nonlinear attenuation of the pump beam and to reabsorption of the signal and test beams.

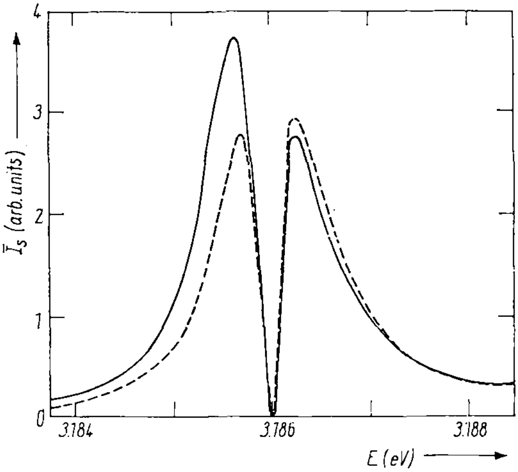

Fig. 6. Excitation spectrum of the (time-integrated) signal emission close to half the biexciton energy ($E_{bi}/2 = 3.186$ eV), using $\hbar\Gamma_{ij} = 10^{-4}$ eV, $\hbar\tilde{\Gamma}_{ii} = \hbar\Gamma_{ii} = 10^{-4}$ eV (dashed line) and $\hbar\tilde{\Gamma}_{ii} = 10^{-4}$ eV, $\hbar\Gamma_{ii} = 10^{-5}$ eV (full line)

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