Phase modulation in second-order nonlinear-optical processes

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If three light pulses interact in a second-order nonlinear-optical process with a phase mismatch, the pulses may acquire a significant phase modulation. This phase modulation leads to a modification of the spectrum of the pulses and may offer the possibility of compressing the pulses. In this paper we discuss the effects of the ratio of intensities of the interacting pulses and the amount of phase mismatch on the phase modulation using analytical and numerical results. When the phase mismatch is small, it turns out that the modulation is also strongly influenced by the initial phase difference between the fields. Finally, we study the combined effects of a phase mismatch and a difference in group velocity on the spectrum and intensity profile of the interacting pulses.

I. INTRODUCTION

A very interesting aspect of nonlinear optics is that it offers a way to modify the frequency spectrum and the time profile of optical pulses. A standard technique to modify the frequency of the light is the application of second-order nonlinear-optical processes such as sum-frequency generation[1–3] (second-harmonic generation) or difference-frequency generation.[4–6]

In order to shorten an optical pulse that is not phase modulated, it is necessary to generate additional frequency components in the spectrum of the pulse. If these new frequency components constitute a linear frequency modulation (linear chirp) over the pulse, this modulation can be compensated by a linear dispersive element and a shortened optical pulse results.

A possible way to generate new frequency components is the application of third-order nonlinear-optical processes such as self-phase modulation (SPM),[7–12] induced-phase modulation (IPM),[13,14] or cross-phase modulation (XPM).[15–23] In all these processes, the refractive index of the medium depends on the intensity of the light. Because the intensity varies over the pulse, the phase velocity varies over the pulse and the pulse acquires a phase modulation while it propagates through the nonlinear medium.

In SPM the phase modulation is caused by the intensity profile of the phase-modulated pulse itself, while in IPM it is caused by the intensity profile of a second copropagating pulse. In IPM the central frequency of the phase-modulated pulse changes if the pulses have a certain delay or if the group velocities are different. XPM is the result of the interference between SPM and some other nonlinear-optical process such as stimulated Raman scattering or second-harmonic generation. It leads to a phase modulation of both the input pulse and the pulse generated by the nonlinear optical process.

A lot of literature deals with the phase modulation that arises as a consequence of third-order nonlinear optical processes.[7–23] In contrast, very little is known about the phase modulation due to second-order nonlinear-optical processes.[24,25]

A primary condition for the generation of a phase modulation in second-order nonlinear-optical processes is that the interaction takes place with a certain amount of phase mismatch. In SPM, IPM, and XPM this is not a necessary condition. In fact, these processes are usually supposed to take place without phase mismatch and are in that aspect simple processes.

The interaction between the electromagnetic fields in second-order processes can be described by three coupled differential equations. These can be solved analytically if the group-velocity differences are negligible. In this description, the amplitudes of the field and the phase difference between the fields are expressed in Jacobi elliptic functions. However, the evolution of the phases of each of the fields individually can only be evaluated by solving an incomplete Jacobi integral of the third kind, which can only be evaluated numerically.

In this paper we discuss the phase modulation occurring in second-order nonlinear-optical processes. In Sec. II we discuss it under the assumption that the interacting pulses have equal group velocities. Section II consists of three subsections. In Sec. II A we derive relations between the phases of the interacting fields. These relations can be graphically depicted and may serve as a qualitative explanation of the phase modulation discussed in Secs. II B, II C, and III.

Sections II B and II C deal with a description of the phase modulation in two limiting cases. In these cases it is assumed that the phase and the amplitude of one of the fields will not be affected by the interaction.

In Sec. III we use a modified Runge-Kutta method[26] in order to evaluate the phase modulation. This section is divided into six subsections. Sections III A–III C are connected with the cases of Sec. II, but no longer use the assumption that one of the fields is unaffected by the interaction. It turns out that the phases of all the fields get modulated and that the modulation is very much dependent on the amount of phase mismatch and the initial ratio of intensities.

Section III D illustrates that if the interaction takes place with a small phase mismatch, the modulation depends on the initial phase difference between the fields.

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Subsections III E and III F deal with the combined effects of phase mismatch and differences in group velocity of the pulses. It turns out that these effects may induce a change in the central frequency and the intensity profile of the pulses.

In Sec. IV we evaluate how the phase modulation that we have calculated in the Secs. II and III is influenced by a change of the parameters of the interaction. We also discuss the differences between the phase modulation generated in second-order processes and the modulation generated in third-order processes.

II. ANALYTICAL

The description of the second-order nonlinear interaction can be greatly simplified if the three interacting fields are assumed to be monochromatic. This is a useful approximation if the spectral bandwidth of the pulses is small compared to their central frequency. The electric component of the three $\mathcal{E}_i$ $(i=1,2,3)$ fields propagating along the z axis can be represented by a product of a field amplitude and a plane wave

$$\mathcal{E}_i(z,t)=E_i(z,t)\exp[i(k_i z-\omega_i t)] , \tag{1}$$

where E_i is the complex amplitude, k_i the wave vector, and ω_i the angular frequency of field $\mathcal{E}_i$.

The interaction results in an energy transfer between the fields. Therefore the field amplitudes are expected to change while they propagate through the nonlinear material. This change in amplitude can be described by three coupled differential equations. These equations are derived from Maxwell's equations and can be simplified to first-order differential equations if the slowly varying amplitude approximation is used. This approximation is valid if the change in amplitude of the fields is significant only after the fields traveled over a distance much longer than their wavelength. The following set of equations results:[27]

$$\left[\frac{\partial}{\partial z}+\frac{1}{v_1^g}\frac{\partial}{\partial t}\right]E_1=\frac{i\omega_1\chi^{(2)}_{\text{eff}}}{2n_1c}E_2^*E_3\exp(i\Delta kz) , \tag{2}$$

$$\left[\frac{\partial}{\partial z}+\frac{1}{v_2^g}\frac{\partial}{\partial t}\right]E_2=\frac{i\omega_2\chi^{(2)}_{\text{eff}}}{2n_2c}E_1^*E_3\exp(i\Delta kz) , \tag{3}$$

$$\left[\frac{\partial}{\partial z}+\frac{1}{v_3^g}\frac{\partial}{\partial t}\right]E_3=\frac{i\omega_3\chi^{(2)}_{\text{eff}}}{2n_3c}E_1E_2\exp(-i\Delta kz) , \tag{4}$$

with v_i^g the group velocity of field $\mathcal{E}_i$, n_i the refractive index of field $\mathcal{E}_i$, $\chi^{(2)}_{\text{eff}}$ the effective nonlinear susceptibility, and Δk the phase mismatch ($\Delta k=k_3-k_2-k_1$).

In order to simplify the interpretation and the numerical evaluation of these equations, the variable t is transformed to η: $\eta=t-z/v_1^g, E_i(z,t)\rightarrow E_i(z,\eta)$ $\{i=1,2\}$ and $E_3(z,t)\exp(i\Delta kz)\rightarrow E_3(z,\eta)$:

$$\frac{\partial}{\partial z}E_1=\frac{i\omega_1\chi^{(2)}_{\text{eff}}}{2n_1c}E_2^*E_3 , \tag{5}$$

$$\left[\frac{\partial}{\partial z}+\left[\frac{1}{v_2^g}-\frac{1}{v_1^g}\right]\frac{\partial}{\partial\eta}\right]E_2=\frac{i\omega_2\chi^{(2)}_{\text{eff}}}{2n_2c}E_1^*E_3 , \tag{6}$$

$$\left[\frac{\partial}{\partial z}+\left[\frac{1}{v_3^g}-\frac{1}{v_1^g}\right]\frac{\partial}{\partial\eta}\right]E_3=\frac{i\omega_3\chi^{(2)}_{\text{eff}}}{2n_2c}E_1E_2+i\Delta kE_3 . \tag{7}$$

The value of $\chi^{(2)}_{\text{eff}}$ is determined by the directions of polarization of the fields and the nonzero elements of the second-rank tensor $\chi^{(2)}$.

When the differences between the group velocities are vanishing, definite relationships between the phases of the complex amplitudes and the phases of the differentials in the complex amplitudes can be derived from the three coupled equations. The following relations hold:

$$\arg(dE_1)=\frac{\pi}{2}-\arg(E_2)+\arg(E_3) , \tag{8}$$

$$\arg(dE_2)=\frac{\pi}{2}-\arg(E_1)+\arg(E_3) , \tag{9}$$

$$\arg(dE_3^i)=\frac{\pi}{2}+\arg(E_1)+\arg(E_2) , \tag{10}$$

$$\arg(dE_3^p)=\frac{\pi}{2}+\arg(E_3) , \tag{11}$$

with $\arg(E_i)$ the phase of the complex number E_i. The differential in E_3 consists of two contributions: one as a result of the nonlinear interaction (dE_3^i) and the other as a result of the phase mismatch (dE_3^p). If there is no interaction, dE_1, dE_2, and dE_3^i vanish.

The phase difference between the differential in complex amplitude and the complex amplitude itself determines whether the amplitude and phase at distance z and time η will increase or decrease. If $|\arg(dE_i)-\arg(E_i)|<\pi/2$, the amplitude of the complex number E_i will increase, in the case $|\arg(dE_i)-\arg(E_i)|>\pi/2$ it will decrease. In the case $0<[\arg(dE_i)-\arg(E_i)]<\pi$ the phase of E_i will increase; in the case $\pi<[\arg(dE_i)-\arg(E_i)]<2\pi$ the phase will decrease. It follows from Eqs. (8) and (9) that

$$[\arg(dE_i)-\arg(E_i)] = \frac{\pi}{2}-\arg(E_1)-\arg(E_2)+\arg(E_3), \quad i=1,2 . \tag{12}$$

The amount of change in amplitude and phase not only depends on parameters that are independent of η such as the phase mismatch, the value of $\chi^{(2)}_{\text{eff}}$, and the frequencies, but also depends on the amplitudes and phases of the E_i themselves. This implies that the E_i at different time points η in the pulse can acquire different phases, with the consequence that the phase over the pulse becomes modulated. This results in a modification of the spectral bandwidth of the interacting pulses.

If the nonlinear optical process takes place leaving one or two of the fields practically unchanged during the in-

teraction, the set of coupled equations can be solved rather easily. In the following we will discuss two examples of these processes. We will derive analytical expressions for the phase modulation as a function of the interaction length. The phase relations between the fields and the differentials of the fields may serve as a qualitative explanation for these expressions.

A. Parametric amplification

In this process an intense field at ω_3 (pump) amplifies two very weak fields at ω_1 and ω_2 (signal and idler). The interaction term in the equation for dE_3 can be neglected because of the relative weakness of signal and idler compared to the pump and the amplitude of E_3 is assumed to be constant. The remaining two equations for E_1 and E_2 can be easily solved. The solution depends on whether the gain factor g_0 of the parametric amplification process is larger or smaller than the phase mismatch Δk.

1. $g_0 > \Delta k$

If $\omega_1=\omega_2$ (and $E_1=E_2$), and assuming that at $z=0$ the phase difference $\Delta\phi=\arg(E_3)-\arg(E_1)-\arg(E_2)$ equals $-\pi/2$, the solution takes the following form:

$$E_1(z,\eta)=E_1(0,\eta)\left[\left[\cosh(\tfrac{1}{2}gz)+\frac{g_0}{g}\sinh(\tfrac{1}{2}gz)\right]^2+\left[\frac{\Delta k}{g}\sinh(\tfrac{1}{2}gz)\right]^2\right]^{1/2}$$
$$\times\exp\left[i\arctan\left[\frac{-\Delta k\sinh(\tfrac{1}{2}gz)}{g\cosh(\tfrac{1}{2}gz)+g_0\sinh(\tfrac{1}{2}gz)}\right]+\tfrac{1}{2}i\Delta kz\right], \tag{13}$$

with $g_0^2(\eta)=\omega_1^2\chi_{\rm eff}^{(2)2}|E_3(\eta)|^2/n_1n_2c^2$ and $g(\eta)=[g_0^2(\eta)-\Delta k^2]^{1/2}$. In this equation the solution for $E_1(z,\eta)$ is written as a complex number with an amplitude and a phase. The amplitude of E_3 is assumed to remain constant so dE_3^i is zero and the solution for $E_3(z,\eta)$ becomes trivial:

$$E_3(z,\eta)=E_3(0,\eta)\exp(i\Delta kz)\ . \tag{14}$$

In the case $gz\gg1$ the terms $\sinh(\frac{1}{2}gz)$ and $\cosh(\frac{1}{2}gz)$ can both be approximated with $\exp(\frac{1}{2}gz)$. The expression for the phase of E_1 becomes

$$\lim_{gz\to\infty}\arg[E_1(z,\eta)]=\arctan\left[\frac{-\Delta k}{g+g_0}\right]+\tfrac{1}{2}\Delta kz+\arg[E_1(0,\eta)]\ . \tag{15}$$

In this limit the phase only depends on z via $\frac{1}{2}\Delta kz$, which implies that the phase modulation that is generated at the beginning of the process [when $\exp(-\frac{1}{2}gz)$ can not be neglected] will remain constant for the rest of the process. It is clear that in this case the phase of E_1^2 which is equal to $2\arg(E_1)$ has the same dependence on z as E_3. This implies that E_1^2 and E_3 rotate through the complex plane with a permanent phase difference equal to $\pi/2+2\arctan[-\Delta k/(g+g_0)]$ [Fig. 1(a)]. In this figure dE_3^p, E_1, and dE_1 are also presented. The phase of dE_1 equals [using Eq. (8)]

$$\arg[dE_1(z,\eta)]=-\arctan\left[\frac{-\Delta k}{g+g_0}\right]+\tfrac{1}{2}\Delta kz+\frac{\pi}{2}+\arg[E_3(0,\eta)]-\arg[E_1(0,\eta)]\ . \tag{16}$$

2. $g_0=\Delta k$

When Δk is equal to $g_0(\eta)$ for a certain value of η, $g(\eta)$ equals zero and the terms $\exp(-\frac{1}{2}gz)$ in the expression for the phase of $E_1(z,\eta)$ can no longer be neglected. In this special case the expression for $E_1(z,\eta)$ becomes

$$\lim_{g\to0}E_1(z,\eta)=E_1(0,\eta)[(1+g_0z+\tfrac{1}{2}g_0^2z^2)]^{1/2}\times\exp\left[\tfrac{1}{2}ig_0z+i\arctan\left[\frac{-g_0z}{2+g_0z}\right]\right]\ . \tag{17}$$

It follows from this expression that for large g_0z the amplitude of E_1 increases linearly with z. The amplitude of dE_1 also grows linearly with z for large g_0z because the amplitude of E_3 was assumed to be independent of z. When z varies from 0 to ∞, the phase of $E_1(z,\eta)$ changes from 0 to $-\pi/4+\frac{1}{2}g_0z+\arg[E_1(0,\eta)]$ [Eq. (17)] and the phase difference between dE_1 and E_1 changes from 0 to $\pi/2$ [Eqs. (12) and (17)], so the orientation of dE_1 with respect to E_1 in the complex plane changes gradually from parallel to perpendicular. The parallel part of dE_1 that changes the amplitude of E_1 is independent of z for large g_0z and remains constant, while the perpendicular part that changes the phase of E_1 increases linearly with z.

In the limit of infinite z the phases of E_1^2 and E_3 become equal to each other [Fig. 1(b)] and E_1 becomes infinitely large. In reality E_3 would deplete if z increases, thus making the assumption that the amplitude of E_3 does not change during the process no longer valid.

3. $g_0<\Delta k$

If Δk is larger than $g_0(\eta)$ the amplification factor $g(\eta)$ becomes imaginary and the amplitude of E_1 will show an oscillatory behavior. Replacing $g(\eta)$ by $ih(\eta)$ in Eq. (7)

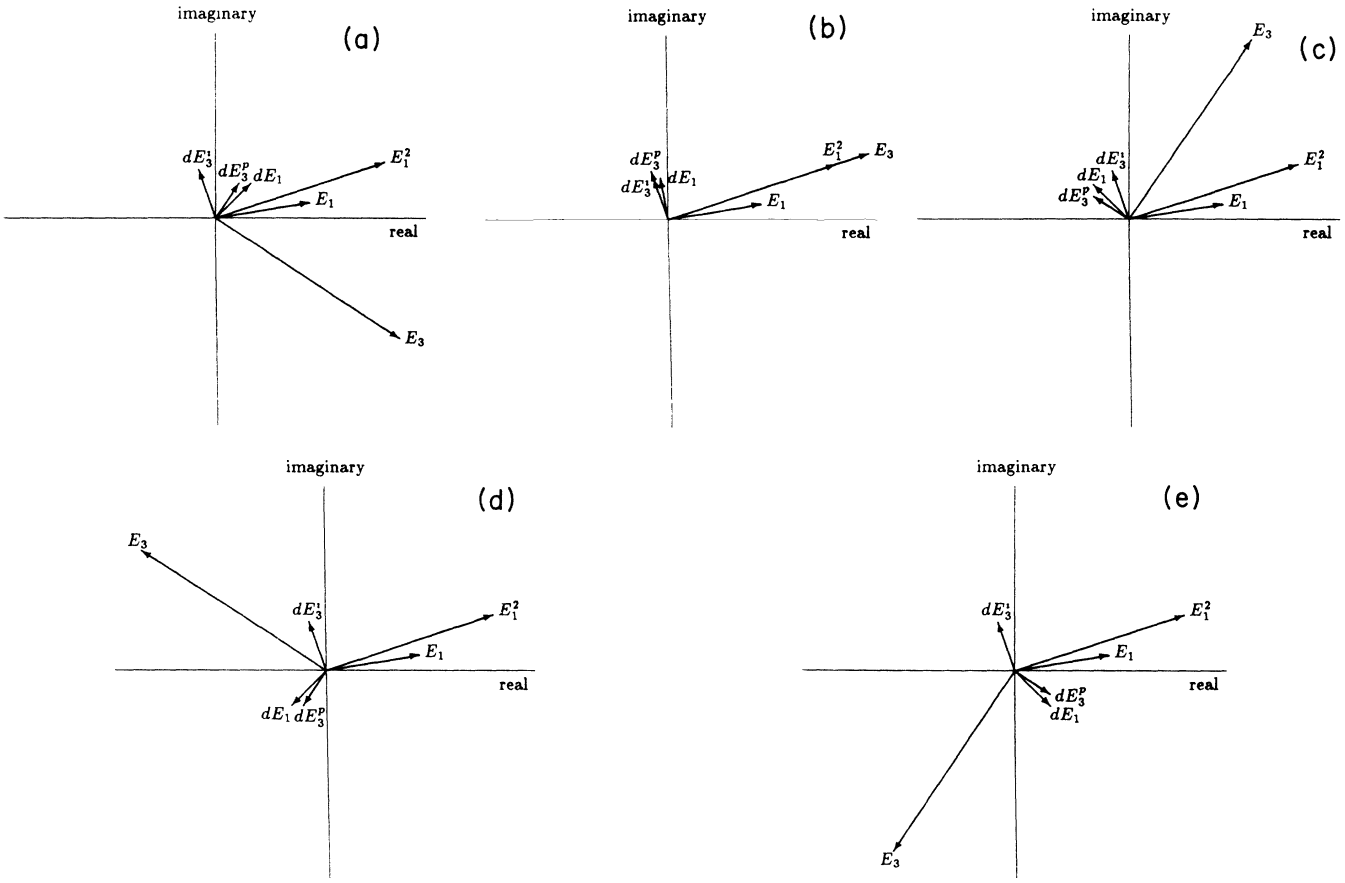

FIG. 1. Orientation of the field amplitudes and differentials in the field amplitudes in the complex plane. The vectors have an arbitrary length.

with $h(\eta)=[\Delta k^2-g_0^2(\eta)]^{1/2}$, the following expression for the phase of $E_1(z,\eta)$ results:

$$\arg[E_1(z,\eta)]=\arctan\left[\frac{-\Delta k\sin(\frac{1}{2}hz)}{h\cos(\frac{1}{2}hz)+g_0\sin(\frac{1}{2}hz)}\right]+\tfrac{1}{2}\Delta kz+\arg[E_1(0,\eta)]\ . \tag{18}$$

The phase of E_1 shows a complicated oscillatory behavior. The phase difference between E_1^2 and E_3 is no longer constant for large z but changes as a function of z. If Δk is positive, the phase of E_1^2 decreases with respect to E_3, when Δk is negative the phase of E_1^2 increases compared to E_3.

The phase difference does not accumulate constantly with z. If Δk is positive, z has to increase with $(2\pi/h)+(2/h)\arctan(-h/g_0)$ in order to decrease the phase of E_1 from 0 to $-\pi/2$ with respect to $\frac{1}{2}\Delta kz$. To decrease the phase further with respect to $\frac{1}{2}\Delta kz$ from $-\pi/2$ to $-\pi$, z has to increase an additional $-(2/h)\arctan(-h/g_0)$.

If $g_0(\eta)$ approaches zero, which is the case in the wings of the E_3 pulse, $(2/h)\arctan(-h/g_0)$ approaches $-\pi/\Delta k$. In that case, the distance z needed for each of the two decreases in phase equals $\pi/\Delta k$. When $g_0(\eta)$ approaches Δk, which is most likely to happen in the center of the E_3 pulse, $h(\eta)$ approaches zero and the distance needed for the first decrease becomes infinitely large. In the same limit $-(2/h)\arctan(-h/g_0)$ approaches $2/g_0$ $(=2/\Delta k)$, and the distance z needed for the first decrease in phase is thus much larger than the one needed for the second decrease in phase. The distances needed to decrease the phase further from $-\pi$ to $-3\pi/2$ and from $-3\pi/2$ to -2π are exactly equal to those of the first and second decrease, respectively.

The difference in the rate of change in phase in the phase intervals can easily be understood if the complex amplitudes and the differentials in the complex amplitude are plotted in the complex plane. It follows from Eq. (12) that if $E_1=E_2$ and $\Delta k>0$, the accumulation is delayed if $-\pi/2<[\arg(E_3)-\arg(E_1^2)]<\pi/2$, which is the case during the first decrease [Figs. 1(a)–1(c), and accelerated if $\pi/2<[\arg(E_3)-\arg(E_1^2)]<3\pi/2$, which is the case during the second decrease [Figs. 1(d) and 1(e)].

The accumulated phase difference depends on η because $h(\eta)$ depends on η. In the center of the pulse of E_3, $g_0(\eta)$ has its maximum value and $h(\eta)$ its minimum. When we compare the rates of change in phase of E_1 in the center and the wings of the E_3 pulse, it can be concluded that in the case $\Delta k>0$, it takes a much longer in-

teraction length in the center to decrease the phase of E_1 from 0 to $-\pi/2$, with respect to $\frac{1}{2}\Delta kz$, than in the wings. For the second decrease from $-\pi/2$ to $-\pi$ it is the other way around, but the difference in distance is much smaller than for the first decrease. The result is that E_1 acquires a higher phase in the center of the E_3 pulse than in the wings. If Δk is negative, it takes a much longer interaction length in the center than in the wings to increase the phase from 0 to $\pi/2$ with respect to $\frac{1}{2}\Delta kz$, so in that case E_1 acquires a lower phase in the center than in the wings.

An expression for the frequency modulation of the E_1 pulse can be derived by differentiating the expression for the phase with respect to $-\eta$ because the frequency is defined as a negative quantity in Eq. (1). If we assume the pump pulse to be a Gaussian in time ($|E_3(\eta)|^2 \sim \exp[-(4\ln 2)\eta^2/\tau^2]$), the following expression results:

$$\Delta\nu_1(z,\eta)=\frac{(\ln 2)\eta g_0^2(\eta)}{\pi\tau^2\Delta k}\left[\frac{z-[1/h(\eta)]\sin[h(\eta)z]}{1-[g_0^2(\eta)/\Delta k^2]\cos[h(\eta)z]+[g_0(\eta)h(\eta)/\Delta k^2]\sin[h(\eta)z]}\right]. \tag{19}$$

If $g_0/\Delta k \ll 1$ and $h(\eta)\gg 1$ for all η (which implies that Δk should be large), the expression simplifies to

$$\Delta\nu_1(z,\eta)=\left[\frac{(\ln 2)\eta g_0^2(\eta)}{\pi\tau^2\Delta k}\right]z\ . \tag{20}$$

It follows from this expression that the frequency modulation takes the form of a linear chirp if $g_0^2(\eta)$ does not vary too much with η, which is the case for the central part of the E_3 pulse.[28] It also follows that the amount of frequency modulation grows linearly with z and that the sign depends on the sign of Δk.

If $g_0(\eta)$ is not much smaller than Δk, it follows from Eq. (19) that there will be peaks in the frequency modulation of the E_1 pulse. These peaks are caused by the fact that if $g_0(\eta)$ approaches Δk, the rate of change in phase of E_1 is very dependent on the phase difference between E_1^2 and E_3. The consequence is that the time profile of the pulse contains intervals in which the phase changes rapidly as a function of z and intervals in which the phase changes slowly as a function of z. This behavior leads to peaks in the derivative of the phase of the pulse with respect to η.

B. Second-harmonic generation

In this process a field at ω_3 is generated or amplified with twice the frequency of an intense fundamental field at ω_1. If E_1 does not change in amplitude or phase as a function of z, the first two of the three coupled differential equations can be neglected and the third can be solved quite easily. In this evaluation we take $E_1(\eta)$ and $E_3(0,\eta)$ real and not phase modulated. Writing the field E_3 as a complex number, the following expression results for the phase:

$$\arg[E_3(z,\eta)]=\arctan\left[\frac{[E_3(0,\eta)+C_3E_1^2(\eta)]\sin(\Delta kz)}{[E_3(0,\eta)+C_3E_1^2(\eta)]\cos(\Delta kz)-C_3E_1^2(\eta)}\right], \tag{21}$$

with $C_3=\omega_3\chi_{\text{eff}}^{(2)}/2n_3c$.

If Δk is positive, the phase of E_3 increases from 0 to $\pi/2$ if z increases from 0 to $(1/\Delta k)\arccos\{C_3E_1^2(\eta)/[E_3(0,\eta)+C_3E_1^2(\eta)]\}$. This distance can vary between 0 if $E_3(0,\eta)=0$ and $\pi/2\Delta k$ if $E_3(0,\eta)$ is infinite. The distance that is needed to further increase the phase from $\pi/2$ to π is equal to $\pi/\Delta k-(1/\Delta k)\arccos\{C_3E_1^2(\eta)/[E_3(0,\eta)+C_3E_1^2(\eta)]\}$ and varies between $\pi/\Delta k$ in the case when $E_3(0,\eta)$ is zero and $\pi/2\Delta k$ when $E_3(0,\eta)$ is infinite, so this distance is always larger than the one needed for the first increase in phase.

This difference in distance needed becomes understandable when the fields and the differentials in the fields are plotted in the complex plane. The differential in E_3 consists of two contributions: dE_3^i due to the interaction and dE_3^p due to the phase mismatch. The orientation of dE_3^i is such that it accelerates the first increase in phase due to dE_3^p [Fig. 1(c)] while it delays the second increase in phase [Fig. 1(d)]. If $E_1(\eta)$ would be small compared to $E_3(0,\eta)$, dE_3^i would be very small compared to dE_3^p and $E_3(z,\eta)$ and would have little effect on the rate of change in phase. In order to increase the phase further to $3\pi/2$ [Fig. 1(e)] would take an additional distance of $\pi/\Delta k-(1/\Delta k)\arccos\{C_3E_1^2(\eta)/[E_3(0,\eta)+C_3E_1^2(\eta)]\}$. This distance is exactly equal to the one needed to increase the phase from $\pi/2$ to π. In order to increase the phase from $3\pi/2$ to 2π [Fig. 1(a)], the same distance is needed as the one needed to increase the phase from 0 to $\pi/2$.

The distances are determined by the argument of the arccos function, which depends on the amplitudes of $E_1^2(\eta)$ and $E_3(0,\eta)$. These amplitudes depend on η, so in principle the argument of the arccos function can be a function of η. If this is the case, the second-harmonic pulse will become phase modulated if $\Delta k\neq 0$. The phase difference between the center and the wings of the pulse,

however, does not accumulate; it oscillates with a period of $2\pi/\Delta k$. Whether the phase of the center will be higher or lower than that of the wings at a certain distance z depends on the amount and the sign of the phase mismatch and the amplitudes and phases of E_1^2 and E_3 at the beginning of the process.

III. NUMERICAL

In this section we will deal with the phase modulation in parametric amplification and second-harmonic generation when all the present field amplitudes and phases are allowed to change as a result of the interaction. If all the interacting fields are intense, it can be expected that the amount of nonlinear interaction, and thus the effect on the phases and the amplitudes of the fields, becomes large. We discuss in this section the influence on the phase modulation of the ratio of the intensities of the interacting fields. We also discuss the effects of the initial phase difference between the fields and the effects of differences in group velocity. The results are obtained numerically using a new modified Runge-Kutta method. The accuracy of this method is of fourth order in both time and distance.[26]

In all examples we assume that the interacting pulses are initially Gaussian shaped and unmodulated and that Δk is positive. We take for the nonlinear material $LiNbO_3$. In all examples, the angular frequency ω_2 is equal to ω_1 and corresponds with a wavelength of 1064 nm. The wavelength corresponding with ω_3 is 532 nm.

A. $E_3 \gg E_1$ and equal group velocities

When the initial phase difference between E_2 and E_1^2 equals $-\pi/2$, the field at ω_3 amplifies the field at ω_1 optimally in the beginning of the process. If this is the case and the phase mismatch is smaller than $g_0(\eta)$, it was shown that the phase difference between E_3 and E_1^2 becomes independent of z.

This is only true under the assumption that the field at ω_3 does not change during the interaction. When the field at ω_3 is initially much more intense than the field at ω_1, this assumption remains valid for a substantial in-

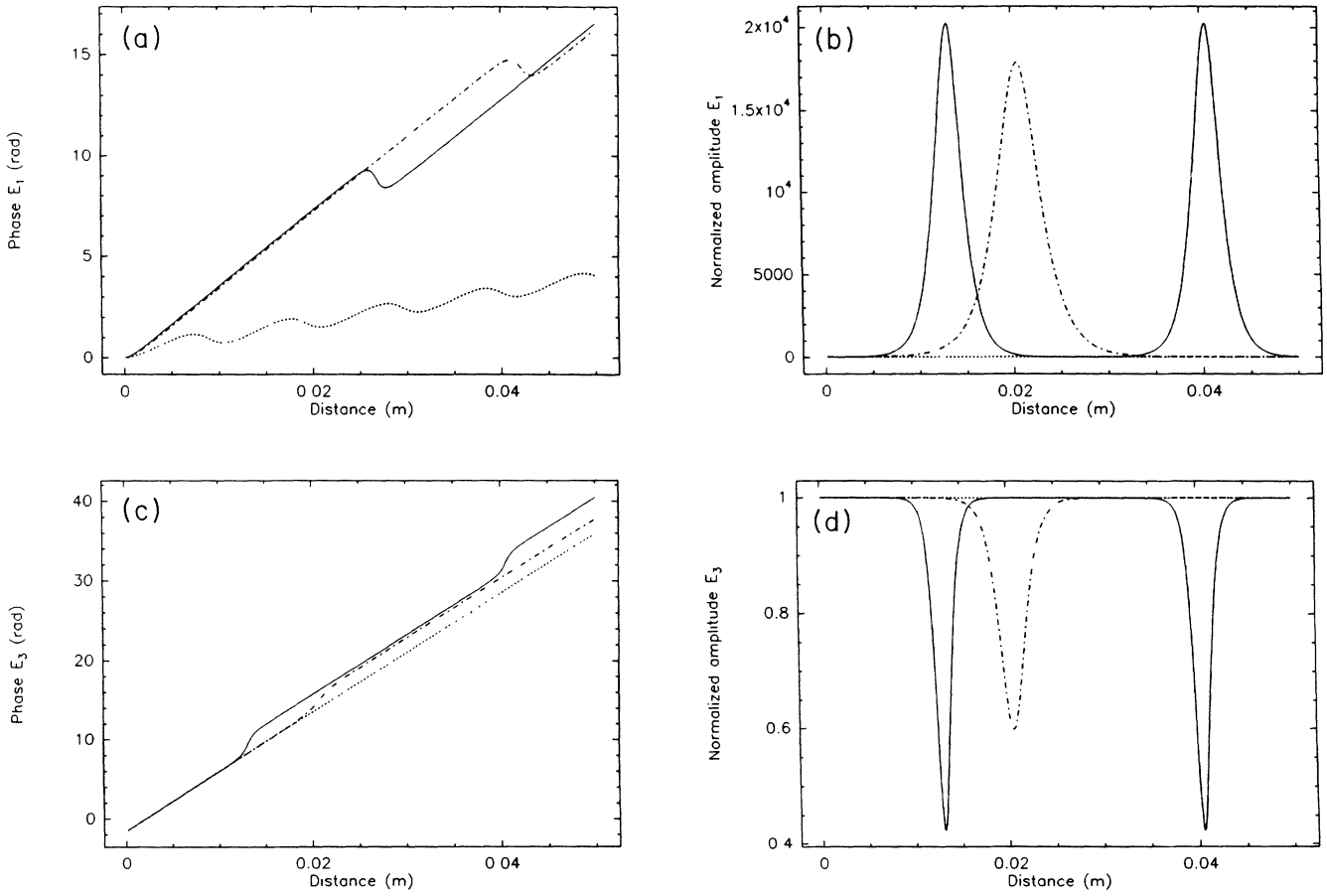

FIG. 2. Calculated phase and amplitude of the field E_1 at fundamental wavelength 1064 nm and the field E_3 at second-harmonic wavelength 532 nm as a function of the interaction length in $LiNbO_3$ for three points in the pulse. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the phase mismatch is 7.5 cm^{-1}. The initial intensities in the centers of the pulses are 1 W/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm (corresponding with an exponential gain factor g_0 in parametric amplification of 17.7 cm^{-1}). The initial phase difference between the fields is $-\pi/2$. ——, at the center of the pulse; — — —, at 5 ps from the center of the pulse; · · ·, at 10 ps from the center of the pulse.

teraction length. After a certain distance, however, the field at ω_3 will get significantly depleted. In that case dE_3^i becomes large with respect to dE_3^p and the phase of E_3 will experience a more rapid increase than the one due to the phase mismatch only. The phase of E_3 becomes larger than that of E_1^2, with the consequence that the orientation of dE_1 with respect to E_1 is such that the amplitude of E_1 is attenuated [Fig. 1(c)]. The phase of E_1 is still increased by dE_1. However, when the phase difference between E_3 and E_1^2 gets larger than $\pi/2, dE_1$ will start to decrease the phase of E_1 [Fig. 1(d)]. The phase of E_1 will now decrease until the phase difference between E_3 and E_1^2 equals $3\pi/2$. At this point the interaction has become similar to the one at the beginning of the process because the phase of E_3 has changed 2π with respect to E_1^2.

The changes in phase and amplitude of the fields E_1 and E_3 as a function of the interaction length are illustrated in Figs. 2(a)–2(d) for three different time points in the pulse. The solid line and the dash-dotted line represent points for which $g_0(\eta) > \Delta k$ and the dotted line represents a point for which $g_0(\eta) < \Delta k$. For the first two points the phase difference between E_1^2 and E_3 only changes because of the depletion of E_3. For the third point the change of the amplitude of E_3 is negligible, but dE_1 is too small to keep the phase difference between E_1^2 and E_3 constant.

In Figs. 3(a) and 3(b) the phase and amplitude of E_1 are presented in the case of $g_0(\eta) < \Delta k$ for all three points in the pulse. It is shown in Fig. 3(a) that in the central point the phase increases for a much longer distance than it decreases. The increases in this figure correspond with the intervals where the phase of E_1 diminishes slowly compared to $\frac{1}{2}\Delta kz$ [Figs. 1(a)–(1c)] and the decreases correspond to intervals where the phase diminishes fast compared to $\frac{1}{2}\Delta kz$ [Figs. 1(d) and 1(e)]. The rates of change in phase of the increases and decreases have approximately the same absolute value because the amplitude of E_3 and thus dE_1/E_1 hardly changes during the process.

In Figs. 4(a)–4(c) the phases, chirps, and intensities of the field at ω_1 are presented as a function of the time over the pulse for three different values of phase mismatch. For all three values of mismatch, the phase in the center is larger than in the wings. The phase over the pulse changes more smoothly as Δk increases. This is caused by the fact that for larger Δk the difference in rate of change in phase at a certain time point η in the pulse between the phase decreases and increases becomes smaller.

The rapid phase changes in the phase profile of the pulse for small Δk lead to peaks in the derivative of the phase with respect to $-\eta$ [Fig. 4(b)]. These frequency peaks, however, will hardly contribute to the spectrum of the pulse because they coincide with points in the pulse with low intensities.

B. $E_3 > E_1$ and equal group velocities

When the intensity of the field at ω_1 is not small at the beginning of the process, the interaction between the fields will be large and the phases and amplitudes of the fields will change rapidly as a function of the interaction distance. In Fig. 5 the evolution of the amplitudes and phases of E_1 and E_3 are presented as a function of interaction length, taking the initial phase difference equal to $-\pi/2$.

We note in Fig. 5(a) that the phase increases much slower in the wings of the pulse than in the situation of Sec. III A. In the center of the pulse the phase of E_1 even decreases. This is caused by the fact that E_3 and thus dE_1 are smaller in the interval in which the phase of E_1 is increased [Figs. 1(a)–1(c)] than in the interval in which the phase of E_1 is decreased [Figs. 1(d) and 1(e)]. In addition, because of the orientation of dE_3, the phase velocity of E_3 is larger in the first interval than in the second, so that this first interval lasts for a shorter distance compared with the situation of Sec. III A, while the second interval lasts for a longer distance. In the center of the

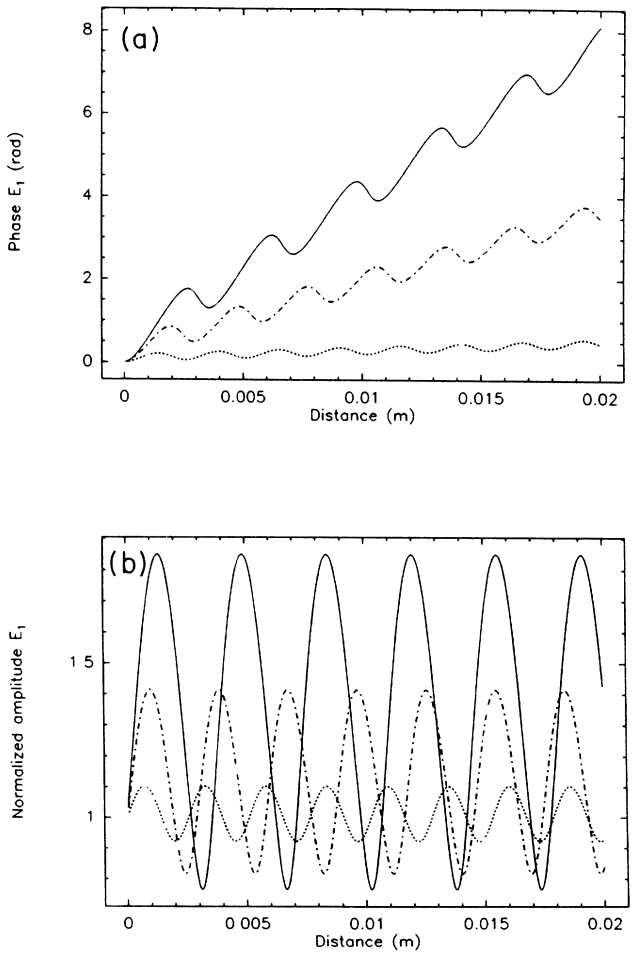

FIG. 3. Calculated phase and amplitude of the field E_1 at 1064 nm as a function of the interaction length in $LiNbO_3$ for three points in the pulse. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the phase mismatch is 25 cm^{-1}. The initial intensities in the centers of the pulses are 1 W/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

pulse where the amplitudes are large, the interaction is larger than in the wings, so that in case of a small Δk, the amplitude difference for E_3 between the intervals can become so large that the net effect on the interaction will be a phase decrease of E_1.

Figure 5(c) shows that the phase of E_3 is also modulated. The phase of the center of the E_3 pulse is larger than the phase of the wings, because for large interaction the interval where dE_3^i increases the phase of E_3 [Figs. 1(a)–1(c)] lasts for a larger interaction length than the interval in which dE_3^i decreases the phase [Figs. 1(d) and 1(e)]. Also the amplitude of dE_3^i is larger (because of the larger E_1^2) in the first interval and has a larger effect because E_3 is smaller in this interval.

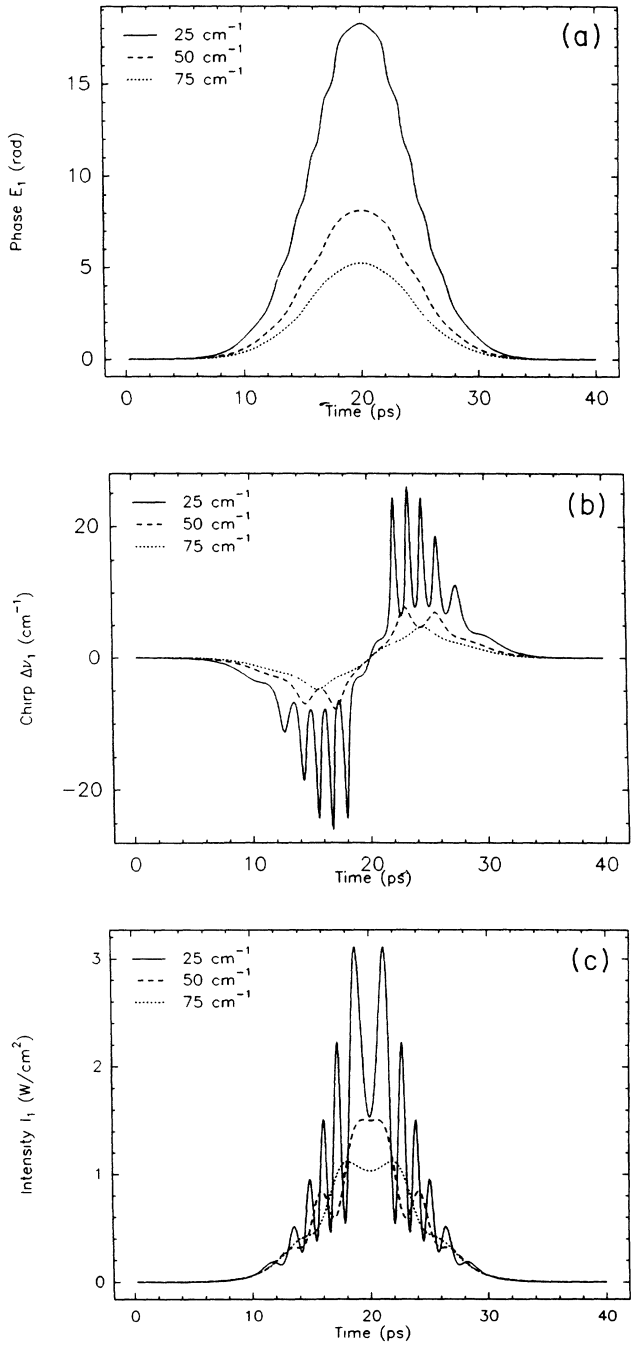

FIG. 4. Calculated phase, chirp, and intensity profile at 1064 nm for three values of phase mismatch after an interaction length of 5 cm $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the initial intensities in the centers of the pulses are 1 W/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

It should be noted that there can only be a net accumulation of phase difference between the center and the wings of the E_3 pulse, in case the amplitude and phase of the E_1 pulse change during the interaction. If E_1 would remain unchanged, it was shown in Sec. II that there will be no net effect.

The profiles of the phases and chirps of the pulses at ω_1 and ω_3 are presented in Fig. 6 for three values of Δk. If the phase mismatch is rather large ($\Delta k > g_0$), the interaction between the fields is similar to the case in which the amplitude of the field at ω_3 is much larger than the one of the field at ω_1. The accumulated phase difference between the center and the wings of the pulse at ω_1 is somewhat smaller than in Sec. III A. The reason for this is given with the description of Fig. 5(a). The frequency chirps on the pulses are of comparable size and are linear for a large part of the pulse, so that compression of the pulse using a dispersive element should be possible.

C. $E_3 \ll E_1$ and equal group velocities

This case is similar to second-harmonic generation. However, in Sec. II B it was assumed that the field at ω_1 does not change during the interaction. If E_3 becomes large during the interaction, due to a large $E_1(0,\eta)$ or a small phase mismatch, this is not a good assumption as illustrated in Fig. 7. The initial phase difference is set equal to $-\pi/2$.

It this case dE_3^i is very large compared to E_3 so that the phase of E_3 increases rapidly in the phase interval in which the phase of E_1 is increased [Figs. 1(a)–1(c)] and slowly in the interval in which the phase of E_1 is decreased [Figs. 1(d) and 1(e)]. The consequence is that the phase of E_3 increases stepwise as a function of the interaction distance [Fig. 7(b)].

Because the rate of change in phase of E_3 is much larger in the first interval than in the second, the interaction distance for the first interval is shorter than for the second, despite the fact that the first interval is lengthened by the increase of the phase of E_1 that takes place in this interval. In addition, the amplitude of E_3 and thus dE_1 is much smaller in the first interval than in the second. This results in a net decrease of the phase of E_1 for all points in the pulse [Fig. 7(a)].

In Fig. 8 the phase, chirp, and intensity profile over the pulse at ω_1 and the phase profile of E_3 are presented for three different values of Δk. The phase profiles and the chirp are rather erratic, especially for small Δk. This is caused by the stepwise phase modulation of E_3, which in turn has its effects on the phase modulation of E_1. The chirp of E_1 has approximately the same value as in Sec. III B, but it is not linear and it has the opposite sign be-

cause the center of the pulse now acquires a lower phase than the wings.

When E_3 is zero at the beginning of the process, Eckardt and Reintjes[24] showed that for the phase of E_3 a rather simple expression results:

$$\arg[E_3(z,\eta)]=\frac{\pi}{2}+\frac{1}{2}\Delta kz+2\arg[E_1(0,\eta)]\ . \qquad (22)$$

This expression is only valid when the amplitude of E_3 does not become equal to zero during the interaction. However, at certain distances the amplitude of E_3 is equal to zero and the phase at E_3 makes a discontinuous jump of π. These distances become smaller as the phase of E_1 decreases. This implies that the phase profile of the pulse at ω_3 will contain several phase jumps of π, dependent on how much the phase of the center of the pulse at ω_1 is lowered compared with that in the wings.

D. Effects of the initial phase difference between E_3 and E_1

In Sec. II it was shown that the initial phase difference between the fields determines whether the energy will flow initially from E_3 to E_1 or vice versa. If the phase mismatch is small, the initial phase difference can have large effects on the evolution of the phase and the amplitude of the pulses. It becomes less important in case of large Δk, because in this case the phase difference between the fields varies rapidly, independent of the interaction.

The effect of changing the initial phase difference is illustrated by comparing Fig. 5 with Fig. 9. In Fig. 5 the initial phase difference is $-\pi/2$, so that at the beginning of the process the field at ω_3 optimally amplifies the field at ω_1. In Fig. 9 the initial phase difference is zero, so that at the beginning of the process neither of the fields is amplified or attenuated and the phase of the fields is maximally changed [Fig. 1(b)].

In this second case (Fig. 9) something special happens: the phase of E_1 increases with approximately half the rate of change in phase of E_3 if $g_0(\eta)>\Delta k$. This can be explained in the following way. The interaction starts with the situation as depicted in Fig. 1(b). In the case when $g_0(\eta)$ is substantially larger than Δk, the phase of E_1^2 will increase more rapidly due to dE_1 than the phase of E_3 due to dE_3^p and dE_3^i. This leads to the situation as depicted in Fig. 1(a). In this situation the amplitude of E_1 increases and the amplitude of E_3 decreases with the result that dE_1 becomes smaller and dE_3 becomes larger.

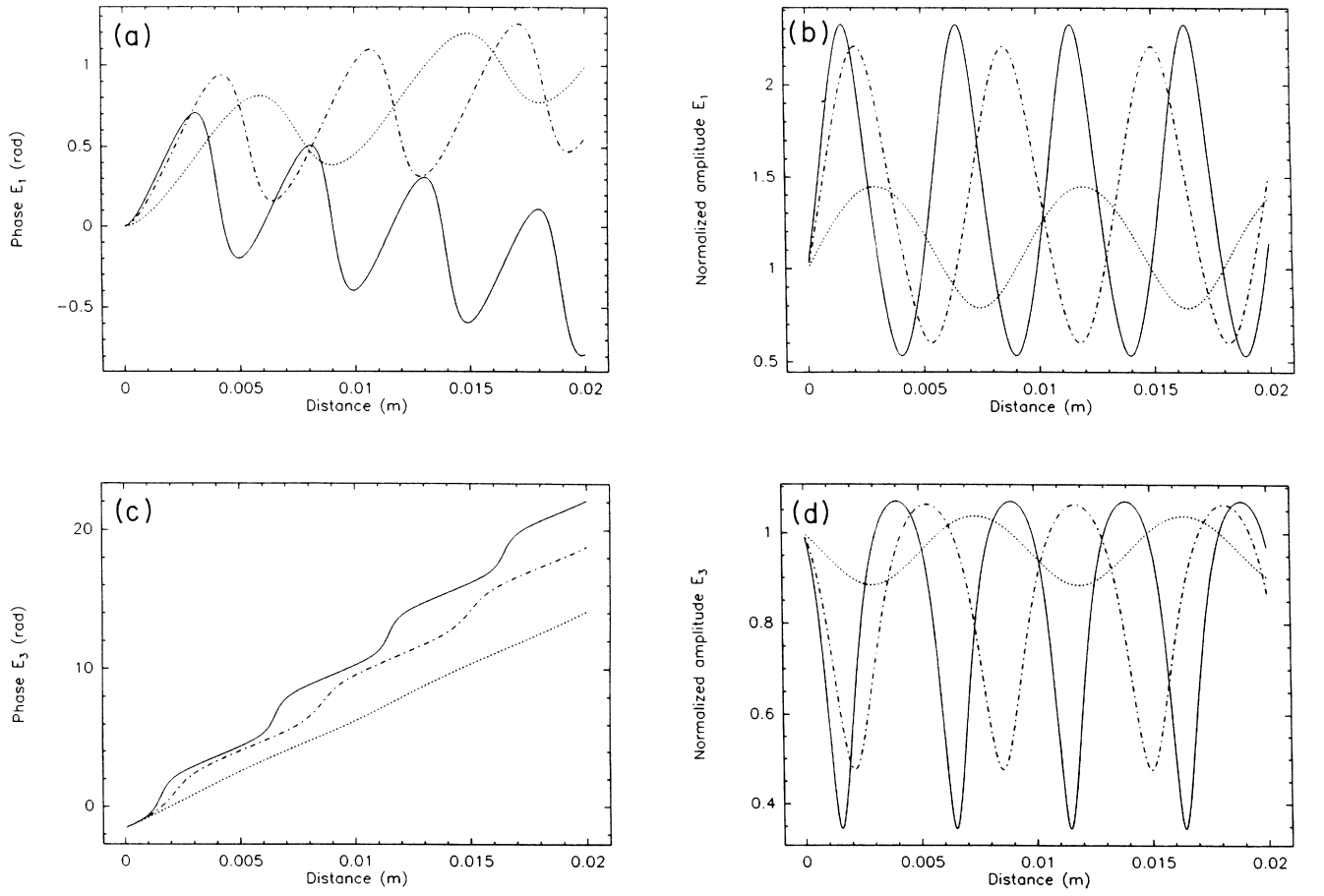

FIG. 5. Same as Fig. 2, but with initial intensities in the center of the pulses of 100 MW/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm.

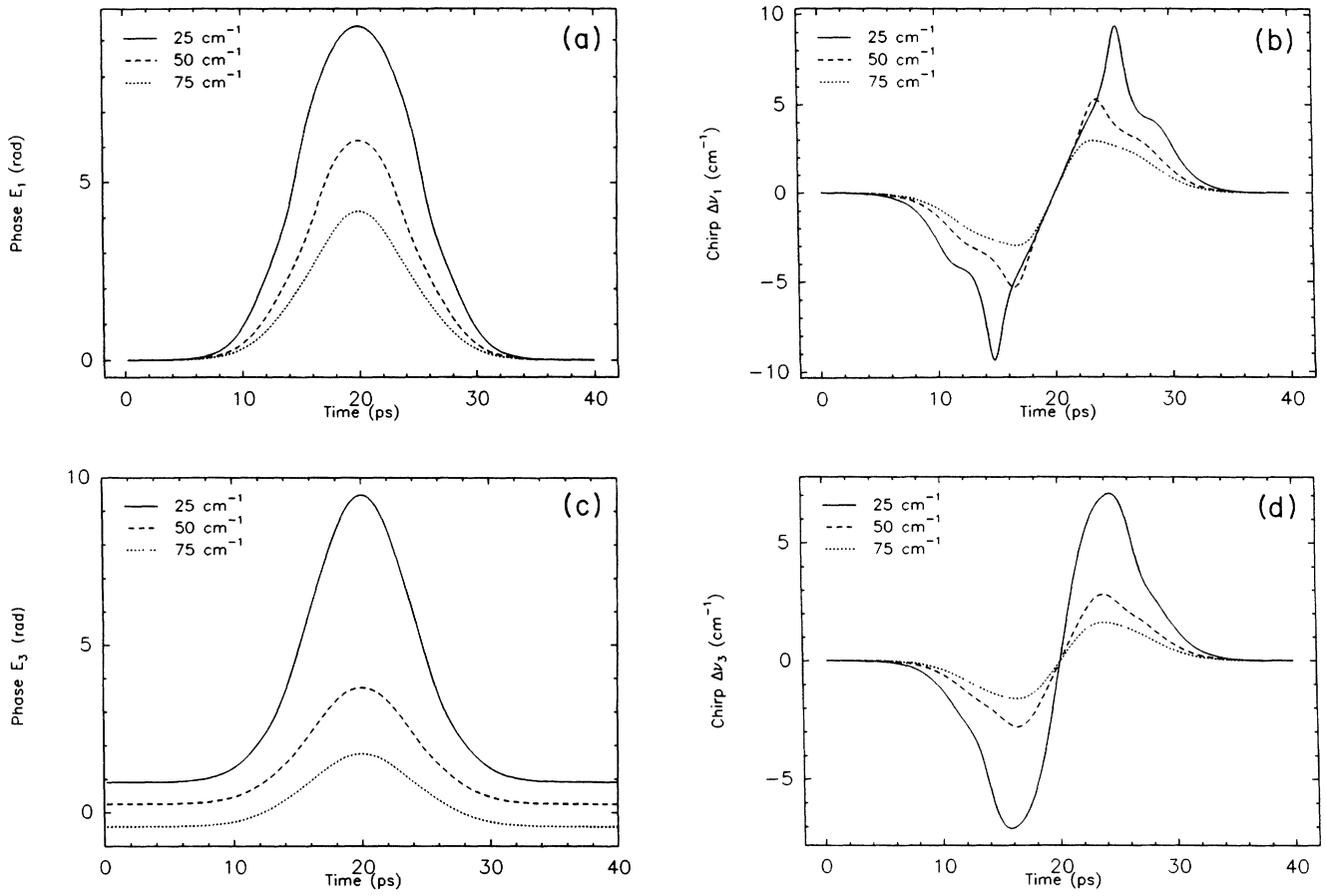

FIG. 6. Calculated phase and chirp profiles of the pulses at 1064 and 532 nm for three values of phase mismatch after an interaction length of 5 cm $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the initial intensities in the centers of the pulses are 100 MW/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

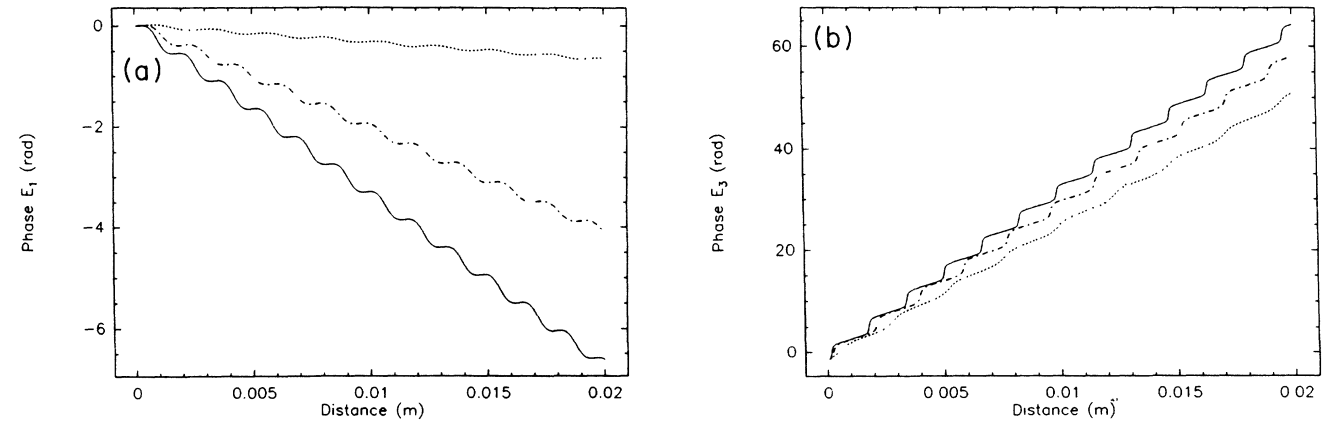

FIG. 7. Calculated phase of the field at 1064 and 532 nm as a function of the interaction length in $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the phase mismatch is 25 cm^{-1}. The initial intensities in the centers of the pulses are 1 GW/cm^2 for the pulse at 1064 nm and 100 MW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

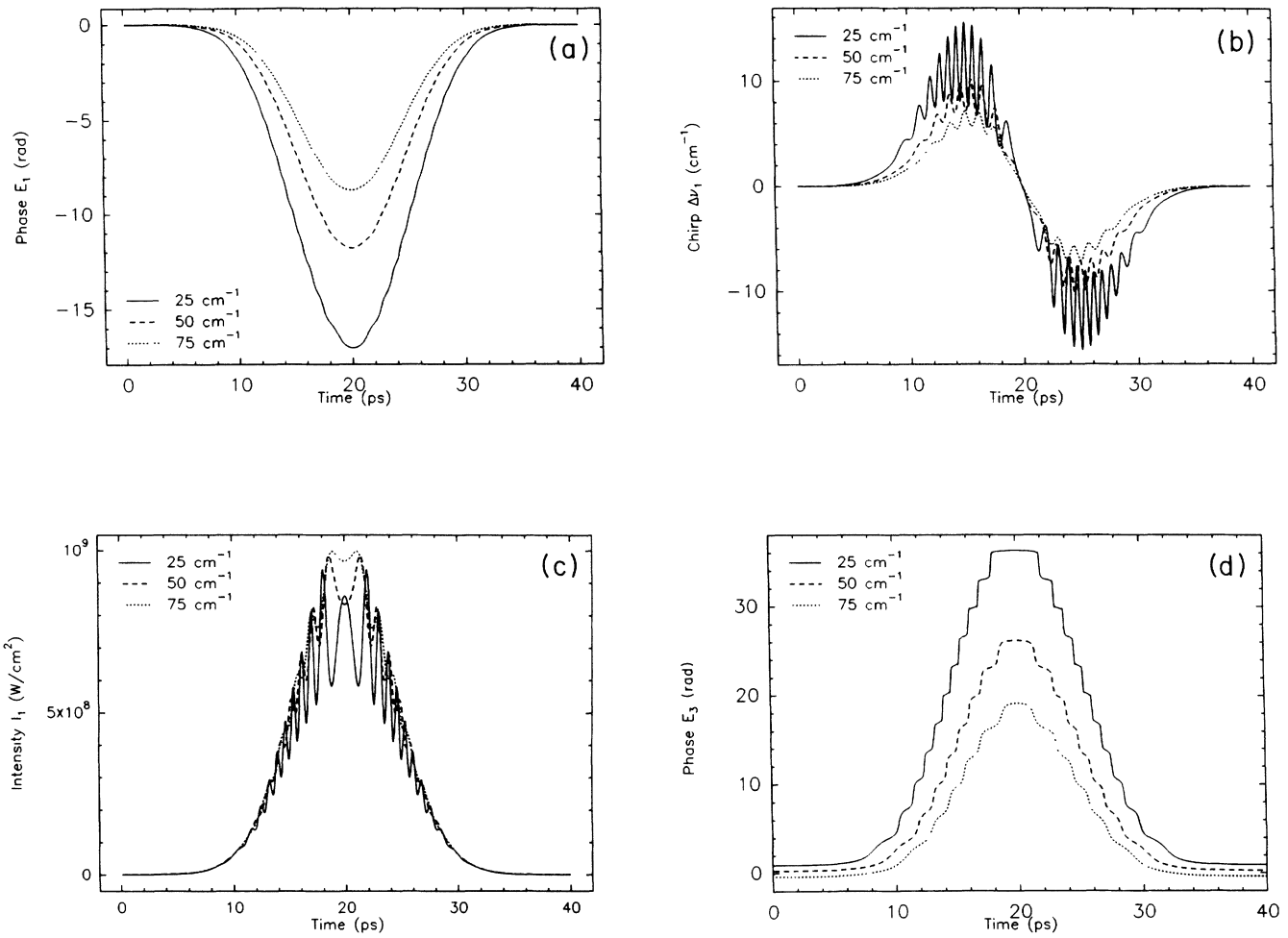

FIG. 8. Calculated phase, chirp, and intensity profile of the pulse at 1064 nm and phase profile for the pulse at 532 nm for three values of phase mismatch after an interaction length of 5 cm $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the initial intensities in the centers of the pulses are 1 GW/cm^2 for the pulse at 1064 nm and 100 MW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

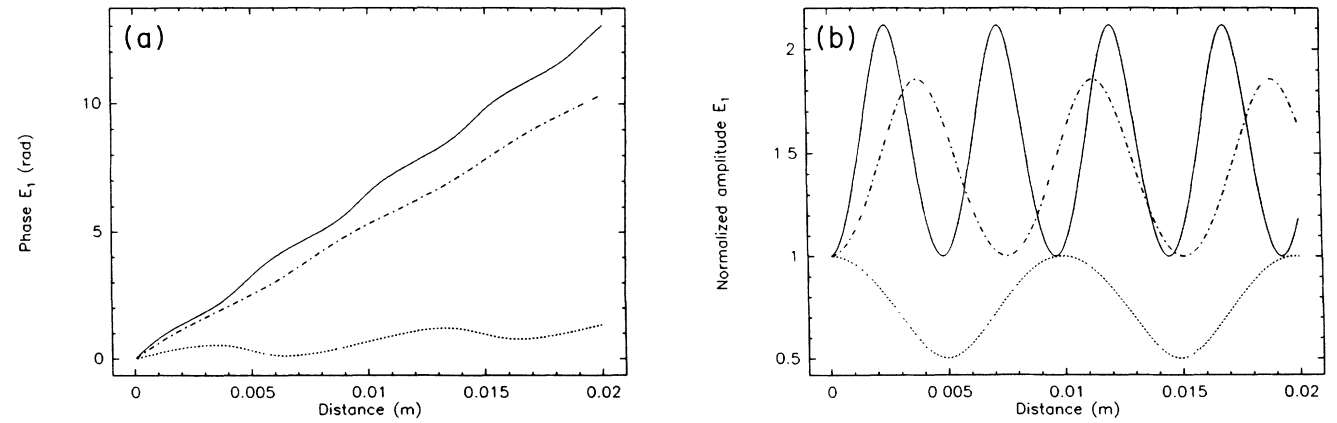

FIG. 9. Same as Figs. 5(a) and 5(b), but with the initial phase difference between the fields equal to zero.

The consequence is that E_3 catches up with E_1^2 and again the situation of Fig. 1(b) arises. Now dE_3 is large and dE_1 small, so that E_3 overtakes E_1^2 and the situation of Fig. 1(c) arises. In this situation the amplitude of E_1 is attenuated and the amplitude of E_3 is amplified so that gradually dE_1 becomes larger and the phases of E_1^2 and E_3 become equal again. In this manner E_1^2 and E_3 are constantly overtaking each other and their phases remain approximately equal for the whole interaction distance.

This phenomenon takes place independently of the amplitude of E_1. It did not occur in the first case (Fig. 5) because the initial phase difference causes E_3 to be very small at the point where the phases of E_1^2 and E_3 become equal to each other. The consequence is that dE_1 cannot become large enough to let the phase of E_1^2 increase with respect to E_3. In this situation the phase of E_3 increases compared to E_1^2 during the whole interaction.

When $g_0 < \Delta k$, which is the case for the point represented by the dotted line, dE_1 is never large enough to keep the phase of E_1^2 up with the phase of E_3, regardless of the initial phase difference. In the case the evolution of the phase is very similar to that of Figs. 3(a) and 5(a).

In Figs. 10(a) and 10(b) the phase profiles of E_1 and E_3 are presented for the two initial phase differences. Special points in these profiles are the points where g_0 is exactly equal to Δk. The phase of E_3 is higher for $\Delta\phi=0$ than for $\Delta\phi=-\pi/2$ because in the first case, the orientation of dE_3 is always such that it increases the phase of E_3, while in the second case there are intervals in which it decreases the phase of E_3.

If $\Delta\phi=0$, the phase difference between the center and the wings is large for both interacting pulses and thus gives rise to a large frequency chirp. This chirp, however, contains large frequency peaks because the phase profile is not very smooth.

When the phase mismatch and the initial phase difference are such that the phase of E_1^2 is increased with respect to E_3 at the beginning of the process, the phases of E_1^2 and E_3 will remain approximately equal during the whole process as described above. It can happen, however, that the phase mismatch and the initial phase difference are such that the phase of E_1^2 would increase with respect to E_3 if only dE_1 and dE_3^p would be taken into account, but if dE_3^i is also taken into account the phase of E_1^2 decreases with respect to E_3. In this case the

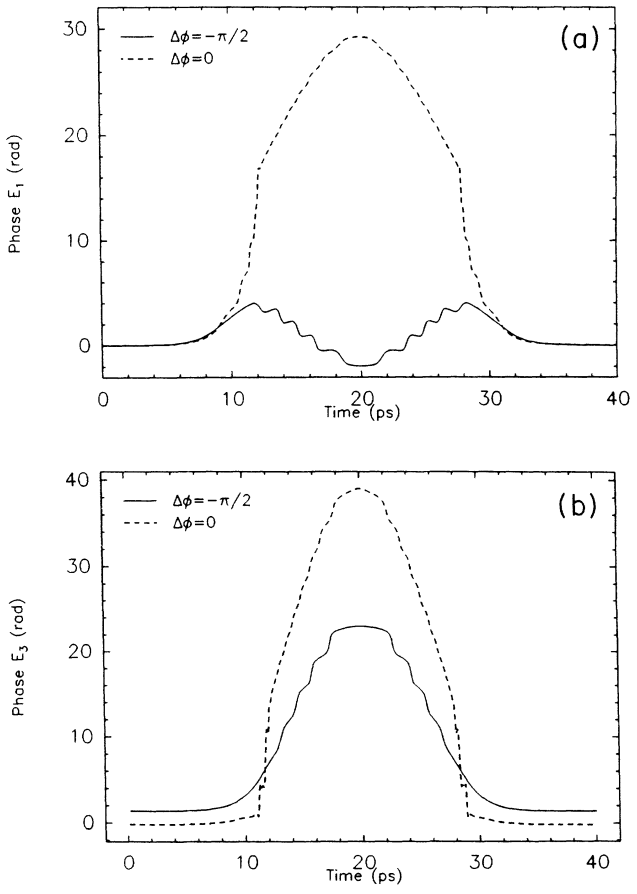

FIG. 10. Calculated phase profiles of the pulses at 1064 and 532 nm for two initial phase differences after an interaction length of 5 cm $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the phase mismatch is 7.5 cm^{-1}. The initial intensities in the centers of the pulses are 100 MW/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm.

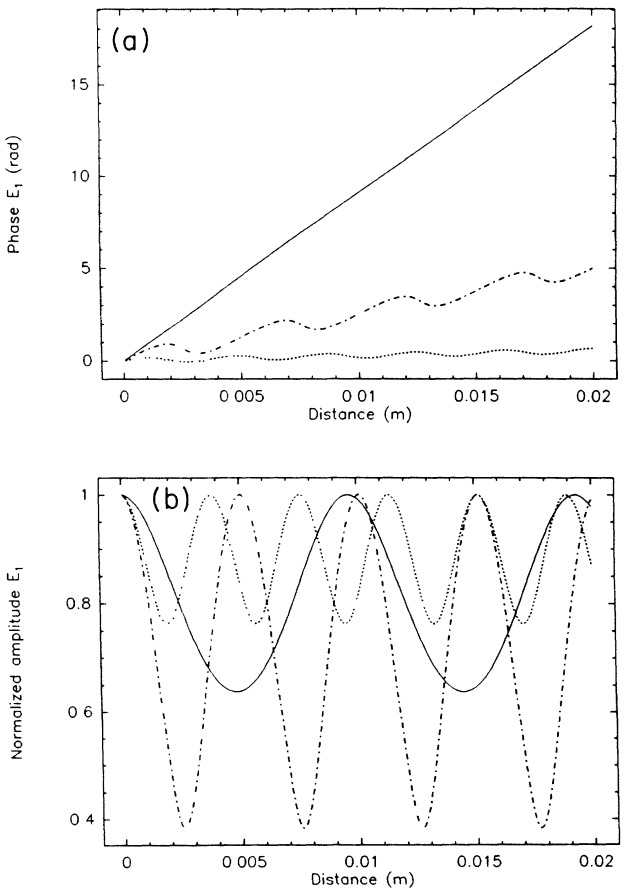

FIG. 11. Same as Fig. 9, but with the phase mismatch equal to 17.0 cm^{-1}.

phases of E_1^2 and E_3 still remain approximately equal, but the process takes place in the opposite direction. If the initial phase difference is zero and Δk is only slightly smaller than $g_0(\eta)$, the phase of E_1^2 will now become initially smaller than E_3 due to the large dE_3^i [Fig. 11(a)], with the result that the amplitude of E_1 initially decreases [Fig. 11(b)].

There can be two points in the pulse for which the change in phase of E_3 due to the phase mismatch and the interaction is exactly equal due to the change in phase of E_1^2 due to the interaction. If the initial phase difference is zero, the amplitudes in these points do not change and the phases of E_1^2 and E_3 remain exactly equal to each other during the whole process.

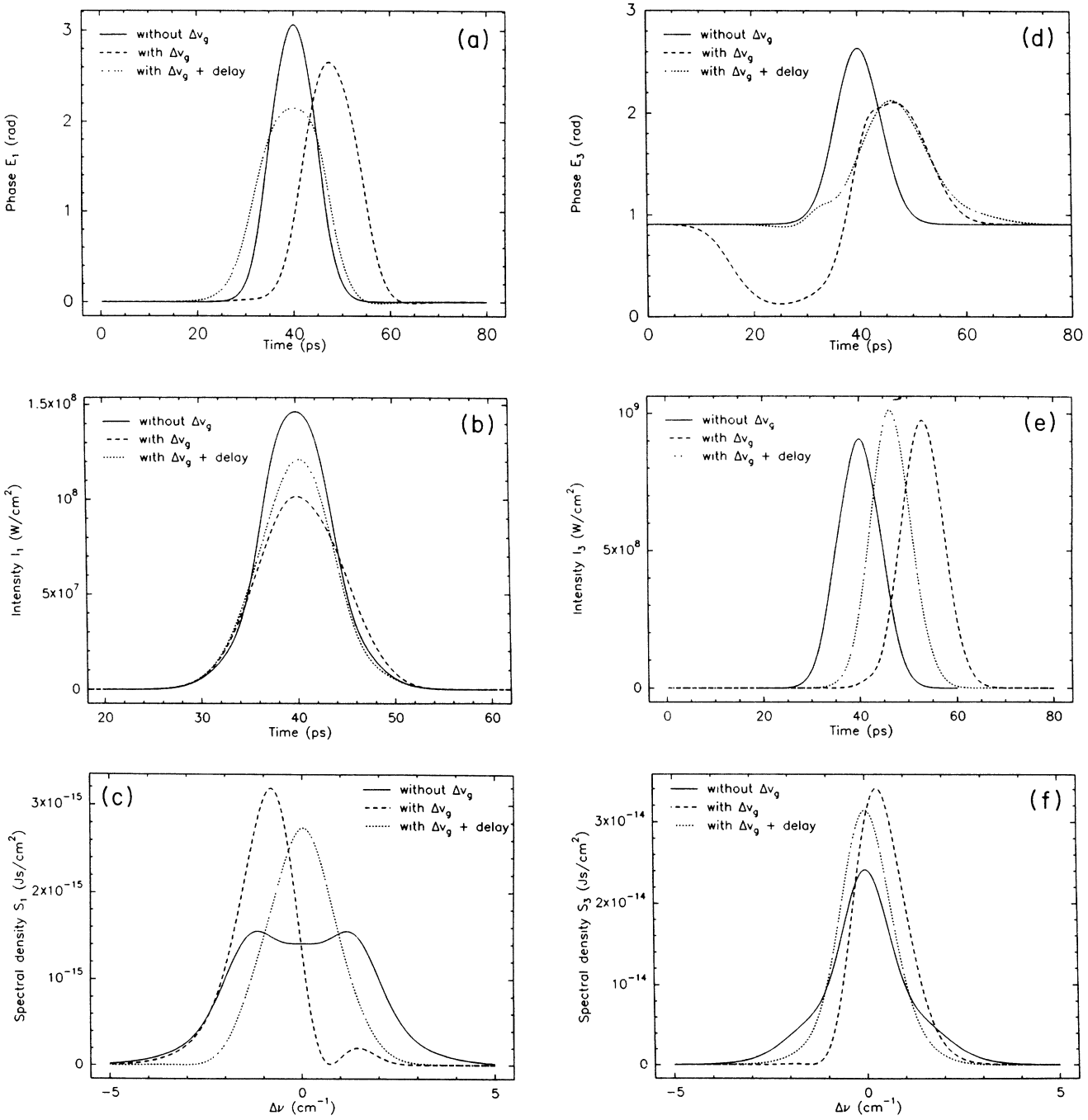

FIG. 12. Calculated phase and intensity profiles and spectra of the pulses at 1064 and 532 nm after an interaction length of 2.5 cm $LiNbO_3$. The initial delay of the pulse at 1064 nm is 6.535 ps in the case represented by the dotted line. The pulse duration of the initially Gaussian-shaped pulses is 10 ps for both pulses and the phase mismatch is 50 cm^{-1}. The initial intensities in the centers of the pulses are 100 MW/cm^2 for the pulse at 1064 nm and 1 GW/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is $-\pi/2$.

E. $E_3 > E_1$ and different group velocities

The phase modulation of the pulses is the result of their interaction. It can be expected that this interaction and thus the modulation changes if the group velocities of the pulses are different. In Fig. 12 the phase and intensity profiles and the spectrum of the interacting pulses are presented if there would be no difference in group velocity, if there are group velocity differences, and if there are group velocity differences and the pulse at ω_1 is initially delayed with exactly half the difference in time the pulses need to travel through 2.5 cm $LiNbO_3$.

In the second case the modulation mainly takes place in the front wing of the slower pulse at ω_3 and the rear wing of the faster pulse at ω_1. This leads to a shift in central frequency of both pulses. The frequency of the pulse at ω_1 will shift to lower frequencies because the phase is higher in the rear wing than in the center and the front wing of the pulse. The frequency of the pulse at ω_3 will shift to higher frequencies because the phase in the front wing is higher than in the rest of the pulse.

In the third case the pulse at ω_3 travels along the pulse at ω_1 during the interaction and is an exactly equal amount of time behind the pulse at ω_1 in the end of the interaction as it was ahead of this pulse in the beginning of the interaction. In this case the phase of E_1 is modulated mostly in the beginning in the front wing, in the end in the rear wing. For the phase of E_3 it is just the other way around. The net effect is that the pulses are mostly modulated in the center and the total phase profiles of the pulses are somewhat broadened compared with the first case in which there is no initial delay and no difference in group velocity. The maxima of the phase modulations will also be lower than in case without group-velocity differences [Figs. 12(a) and 12(d)], so the pulses will be less spectrally broadened [Figs. 12(c) and 12(f)].

F. $E_3 \ll E_1$ and different group velocities

This process has been studied previously by several authors.[19,26,29,30] It has been shown that if the difference in time the pulses need to travel through the nonlinear material exceeds the pulse duration of the pulse at ω_1 and the process takes place with a significant phase mismatch, a two-peak structure for the pulse at ω_3 results. This structure is the result of the fact that the light that is gen-

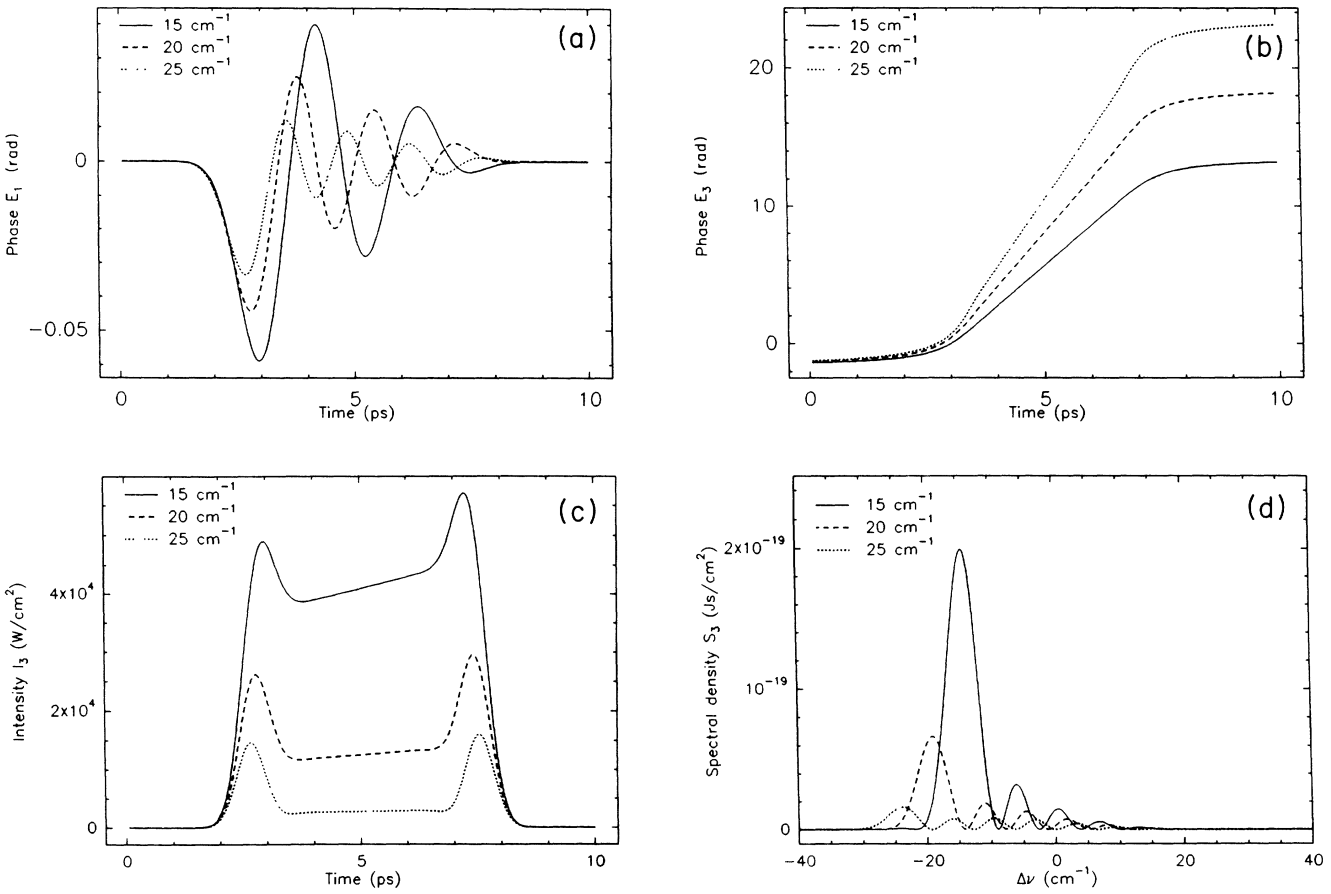

FIG. 13. Calculated phase profiles of the pulses at 1064 and 532 nm and intensity profile and spectrum of the pulse at 532 nm for three values of phase mismatch after an interaction length of 1 cm $LiNbO_3$. The pulse duration of the initially Gaussian-shaped pulse at 1064 nm is 1 ps and the initial intensities in the center of the pulses are 4 MW/cm^2 for the pulse at 1064 nm and 0 W/cm^2 for the pulse at 532 nm. The initial phase difference between the fields is equal to $-\pi/2$.

erated in the front wing of the pulse at ω_1 destructively interferes with the light that is generated in the rear wing. The final pulse will thus mainly consist of light that is generated in the rear wing at the beginning of the process and light that is generated in the front wing at the end. In Fig. 13 the phase profiles of E_1 and E_3, and the intensity profile and the spectrum of the pulse at ω_3, are presented for three values of phase mismatch. The phase of E_3 increases rapidly over the two-peak structure. This increase is caused by the fact that the light in the rear part of the pulse was generated at the beginning of the process and has accumulated a higher phase due to the positive phase mismatch than the light that has just been generated at the front part of the pulse.

For higher values of phase mismatch this accumulated phase difference is also higher. The phase difference and the delay between the two peaks can be set by varying the phase mismatch and the interaction length. This implies that the two-peak structure might be used in phase-sensitive pump-probe experiments.

The phase modulation of the pulse at ω_1 is a direct consequence of the phase modulation of E_3. The phase difference between E_3 and E_1 determines whether the phase of E_1 is increased or decreased. For higher values of phase mismatch, the phase of E_3 passes through more phase intervals and the phase profile of E_1 acquires more oscillations. The central frequency of the pulse at ω_3 is shifted because the phase of E_3 increases over the pulse. The spectrum of this pulse consists of fringes (Ramsey fringes), which is caused by the fact that the pulse mainly consists of two peaks. The spacing of the fringes is defined by the inverse of the time delay between the two peaks.

IV. DISCUSSION

The analytical and numerical results show that the phase modulation in second-order nonlinear-optical processes depends on the phase mismatch, the initial phase difference, and the parameters of the incoming fields. Of these parameters, the intensities of the pulses ω_1 and ω_3, and especially the ratio of these intensities, are very important for the character of the modulation. The pulse duration is only important for the phase modulation if there is a significant difference in group velocity between the interacting fields. If the group velocities are equal, the phase difference between the center and the wings of the pulse is independent of the pulse duration. The consequence is that the factor by which the bandwidth is increased does not depend on the pulse duration, in spite of the fact that the chirp and the spectral bandwidth of the pulses are both inversely proportional to the pulse duration.

If the group velocities are equal, the phase modulation can easily be scaled by multiplying the coupling terms of the fields on the right-hand side of Eqs. (5)–(7) with the same factor. This implies that all electric fields and the phase mismatch should be multiplied with the same value. If the interaction distance is multiplied with the inverse of this scaling factor, the same phase modulation results as before scaling.

If the group velocities are different, the phase modulation is no longer independent of the pulse duration and the pulse duration should also be multiplied with the inverse of the scaling factor in order to obtain the same relative delay between the pulses during the interaction as before scaling. Like the ratio of intensities, the influence of the initial phase difference on the phase modulation cannot be scaled. However, this phase difference is only crucial for the character of the modulation if the phase mismatch is smaller than the exponential gain factor g_0 defined for parametric amplification.

The phase modulation generated in second-order nonlinear-optical processes differs in many aspects from that generated with third-order processes. In the first place, the amount of phase modulation in a second-order process like parametric amplification scales with the square root of the intensity of the strongest pulse, while in SPM and IPM it scales linearly with intensity.

Second, the amount and sign of the modulation can very easily be tuned in second-order processes by rotating the nonlinear crystal. In a third-order process, the amount of modulation can only be tuned by varying the intensity or the interaction length and a positive modulation can only be changed in a negative one by replacing the nonlinear material for an absorbing medium in which the nonlinear refractive index can be negative.

In the third place the pulse modulates its own phase in SPM whereas in IPM and in second-order processes the phase is modulated by the interaction with another pulse. The advantage of SPM is that the modulation is not influenced by a difference in group velocity between the pulses, so that even for very short pulses the phase can be modulated over a long interaction length. We showed in Sec. II E that this problem can partly be circumvented by giving the fastest pulse an initial delay so that this pulse is an equal amount of time ahead of the slowest pulse at the end of the crystal as it was behind in the beginning. However, the difference in group velocity between the pulses limits the interaction length in which the pulses overlap and thus poses an upper limit to the amount of phase modulation.

In the fourth place, second-order processes can be efficient in generating a phase modulation with relatively low intensities and short interaction lengths because the $\chi^{(2)}$ of nonlinear crystals is in general much higher than the nonlinear refractive index. For $LiNbO_3$, the value of $\chi^{(2)}_{\text{eff}}$ is equal to 1.16×10^{-11} m/V (Ref. 27) when the polarization is taken parallel to the optical axis. The nonlinear refractive index of $LiNbO_3$ is estimated to be 1×10^{-13} esu$=1.39\times10^{-21}$ m^2/V^2.[31] In order to compare the third-order modulation with the second-order modulation, we calculate the phase modulation of a 1-GW pulse at 532 nm in a 5-cm $LiNbO_3$ crystal due to the nonlinear refractive index with the following equation:[32]

$$d\phi=\frac{\omega n_2 IL}{4\pi\epsilon_0 c^2 n_0}\ , \tag{23}$$

with n_2 the nonlinear refractive index, I the intensity, L the interaction length, $1/4\pi\epsilon_0=9\times10^9$, c the velocity of light in vacuum, and n_0 the extraordinary refractive in-

dex of $LiNbO_3$ (equal to 2.2345). It follows that the phase is modulated with 0.11 rad, which is in general negligible to the modulation of the phase in a phase-mismatched second-order process.

Considering these differences between second-order and third-order processes, we conclude that second-order processes are very useful in experiments in which the generated phase modulation should be tunable and in experiments in which the intensity is rather low. For very short pulses and high intensities, SPM is more useful because this process is not influenced by group velocity effects and the phase modulation increases linearly with intensity.

V. CONCLUSION

In this paper we have shown that second-order nonlinear-optical processes may lead to a phase modulation of the interacting pulses. This phase modulation can be qualitatively explained when the fields and the differentials in the fields are plotted in the complex plane.

It turns out that if the processes take place with a phase mismatch, the interaction leads in almost any case to an accumulation of phase difference between different parts of the pulse. Only in the case where the field at angular frequency ω_1 does not change due to the interaction, no accumulation of phase difference occurs. When the field at ω_3 is more intense than the one at ω_1, the interaction leads to a linear chirp on both pulses.

If the phase mismatch is smaller than the exponential gain factor g_0 defined for parametric amplification and the initial phase difference between the fields is small, the phase modulation can become very large and the amount of phase modulation of the field at ω_1 is approximately half the amount of the field at ω_3. This implies that in the center of the pulse the phase mismatch is fully compensated by the interaction.

If the group velocities of the interacting pulses are different, the phase modulation leads to a change in central frequency. This shift can be avoided by giving the pulses an initial delay. If the intensity of the field at ω_3 is initially small and the difference in group velocity is large, the combined effects of phase mismatch and difference in group velocity lead to the generation of a two peak structure for the field at ω_3. The phase difference between the two peaks depends on the phase mismatch and the interaction length. The sign and amount of the phase modulation in second-order optical processes can be easily tuned by rotating the nonlinear crystal and the modulation can be generated with relatively low intensities and short interaction lengths compared to third-order processes.

Finally, we conclude that second-order nonlinear optical processes offer a useful tool to modulate the phase of short optical pulses. This modulation may be used for experimental applications in pulse compression, frequency conversion, and phase-sensitive pump-probe experiments.

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