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Photonics in quadratic nonlinear media beyond the $\chi^{(3)}$ perspective

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We fully analytically describe the amplitude and phase modulation which arise due to cascaded second-order interaction. We show that an interplay between nonlinearity and mismatch determines the characteristic features of the output modulation. Simple equations for the phase modulation in the limiting case of moderate mismatch are derived. For the nonstationary situation conditions are identified where a homogeneous phase shift across the pulse can be accomplished being very important for the performance of phase-sensitive switching devices such as nonlinear Mach–Zehnder interferometer or loop mirrors.

1. Introduction

For more than a decade, degenerated cubic nonlinear effects have been preferred in all-optical processing schemes (see, *e.g.*, for an overview [1]). In this cubic scenario power- and distance-dependent self-phase modulation of one wave by itself and cross-phase modulation of two waves are the basic effects. As far as all-optical switching is concerned the phase modulation can only be transformed into an amplitude modulation in particular devices being sensitive to phase changes as interferometers (Mach–Zehnder, Fabry–Perot, Sagnac) or directional couplers. A serious obstacle to the implementation of such all-optical switching devices based on cubic nonlinearities and operating at reasonable power levels are the small off-resonant Kerr nonlinearities. They are even small in such promising materials as direct semiconductors near half-the-band-gap energy [1]. In a search for alternatives quadratic nonlinear effects were revised recently bringing back to mind the well-known fact that a nonlinear phase modulation can also arise in this situation [2]. Due to the cascaded up- and down-conversion upon propagation this modulation is acquired by all waves injected. Hence, it can be used for an all-optical manipulation of the fundamental frequency (FF) signal as in a cubic nonlinear environment [3]–[7]. With regard to all-optical switching the attention should not be merely focused on a search for effects known from cubic materials. The essential peculiarity of the quadratic effects is the unavoidable coexistence of phase- and amplitude modulation. Hence, on the one hand, this is advantageous for switching because no phase sensitive devices are required. On the other hand, this might be detrimental if phase modulation is intended to be exploited because the amplitude modulation appears as an undesired side effect.

Early studies [5]–[7] were mostly concentrated on the limit of large wave vector mismatch. In this particular situation, it has been numerically predicted [7] and experimentally shown [8] that the phase modulation of the FF wave is compared qualitatively to that emerging in a cubic nonlinear material and that the amplitude modulation is only minor. Although this case only covers a very restricted experimental situation, it tells us that cascaded second-order processes can be potentially more effective than third-order ones, provided that the extremely large second-order nonlinearities of semiconductors or poled polymers can be exploited. Evidently, the general case of arbitrary mismatch yields a more complicated evolution of the phase and, moreover, an unavoidable combination of phase and amplitude modulation.

The aim of this paper is to study quantitatively amplitude and phase modulation in cascaded interaction in a quadratic medium considering the effect of the phase mismatch, to analyse switching prospects, and to compare them with the traditional cubic schemes. The paper is organised as follows: In Sect. 2 we briefly review basic equations and their solutions for both amplitude and phase modulation. Formulas for the effective nonlinear cubic coefficient in different approximations are derived in Sect. 3. Peculiarities of the output modulation in the nonstationary situation are discussed in Sect. 4. A short summary concludes the paper.

2. Analytical description of wave interaction

In what follows we concentrate on the so-called vectorial or type II interaction, where two orthogonally polarised FF waves a_j interact with the second harmonic (SH) wave b. The evolution of three waves is described by a coupled set of equations [9]:

$$\frac{da_j}{dZ} = i\gamma a^*_{3-j} b, \quad \frac{db}{dZ} = i(\gamma a_1 a_2 - 2kb), \quad j = 1, 2 \tag{1}$$

where normalised quantities for the propagation distance, the mismatch and the amplitudes are introduced as:

$$Z = z/L, \quad k = \Delta\beta L/2, \quad a_j = \sqrt{2}A_j/\sqrt{P}, \quad b = (B/\sqrt{P})\exp(-i\Delta\beta z).$$

Here L is the device length and $P = |A_1|^2 + |A_2|^2 + |B|^2$ is the total guided power or the total intensity of the plane waves in the bulk crystal, respectively. The wavevector mismatch is defined as $\Delta\beta = \beta_1(\omega) + \beta_2(\omega) - \beta(2\omega) + g$ with $g = 2\pi/p$ and p is the periodicity of the grating optionally imposed to get quasi-phase matching [10]. The crucial quantity describing the effectivity of the nonlinear conversion and playing a key role in the subsequent discussion is defined as $\gamma = L\chi_{\text{eff}}\sqrt{P}$ (χ_{eff} is the effective second-order nonlinear coefficient) ans is henceforth termed as nonlinearity. In the nonstationary case the parameter γ depends on time τ according to the pulse shape (instantaneous amplitude $\propto \sqrt{P}$).

The system (1) can be straightforwardly integrated [11]. Provided that the group velocity mismatch is negligible for zero initial SH input, we get the evolution of FF profiles as

$$|a_{1,2}(Z,\tau)|^2 = (1\pm\delta)p(\tau)-u_-(\tau)\mathrm{sn}^2(\sqrt{u_+(\tau)}\gamma Z|m(\tau)), \tag{2}$$

$$u_\pm(\tau) = \frac{1}{2\gamma^2}\left(2\gamma^2 p(\tau)+k^2\pm\sqrt{k^4+4\gamma^2 p(\tau)[k^2+p(\tau)\gamma^2\delta^2]}\right), \tag{3}$$

$$m(\tau) = u_-(\tau)/u_+(\tau). \tag{4}$$

Here, for the weaker a_1 and the stronger a_2 FF waves the $-/+$ sign holds, respectively, $p(\tau)$ represents the pulse shape supposed for clarity to be Gaussian $[p(\tau) = p_0\exp(-\tau^2)]$ and to have the same width for any input wave, *i.e.*, the parameter δ represents the input imbalance between the two FF components $(|a_1(0,\tau)|/|a_2(0,\tau|)^2 = (1-\delta)/(1+\delta)$. The evolution of the phase profile of the weaker and the stronger FF envelopes can also be obtained integrating (1) [12]:

$$\varphi_\pm(Z,\tau) = -kZ+\frac{(k/\gamma)}{\sqrt{u_+(\tau)}}\Pi\left[n_\pm;\mathrm{am}(\sqrt{u_+(\tau)}\gamma Z|m(\tau))|m(\tau)\right], \tag{5}$$

$$n_\pm(\tau) = \frac{u_-(\tau)}{(1\pm\delta)p(\tau)} \tag{6}$$

where $\Pi(u;\vartheta|m)$ is the elliptic integral of the third kind, $\mathrm{am}(v|m)$ is the Jacobi amplitude function, and $u_\pm$ and m are given by (3) and (4), respectively. Using Eqs. (5) and (6), the optimum ratio mismatch/nonlinearity can be calculated to obtain phase shift required for the operation of various all-optical schemes [12].

Frequently, one encounters situations where the linear mismatch $|k|$ exceeds the nonlinearity γ. We found that the fairly involved Eq. (5) describing the evolution of the phases can be considerably simplified, where $|k| \gg \gamma$ is not required. By inspecting the parameter of the elliptic functions m as a function of $|k|/\gamma$ it turns out that this parameter tends to zero as $|k|/\gamma$ grows. Hence, the elliptic integral of the third kind can be approximated by trigonometric functions $\Pi(n;\vartheta|m) \approx \arctan(\sqrt{1-n}\tan\vartheta)/\sqrt{1-n}$. Note that the domains where the function $\Pi(n;\vartheta|m)$ and $\arctan\vartheta$ are defined do not coincide, which requires particular care to derive the exact relation. In doing so one gets a simple relation for the phase shift where only trigonometric rather than elliptic functions are involved. Note that it suffices to require $|k|/\gamma \gtrsim 1$ to get instead of (5)

$$\varphi_\pm(Z,\tau) = -kZ+\left(\frac{k}{\gamma}\right)\frac{G[\vartheta(Z,\tau)]+\pi[\mathrm{int}(2\vartheta(Z,\tau)/\pi)-\mathrm{int}(\vartheta(Z,\tau)/\pi)]}{\sqrt{u_+(\tau)[1-n_\pm(\tau)]}}. \tag{7}$$

Here $\vartheta(Z,\tau) = \sqrt{u_+(\tau)}\gamma Z$, $G(x) = \arctan\left(\sqrt{1-n_\pm}\tan x\right)$ and $\mathrm{int}(x)$ denotes the integer part of x. In Figure 1, the results of an exact calculation (5), (6) are shown for different ratios $|k|/\gamma$. An approximate solution obtained from (7) is plotted for

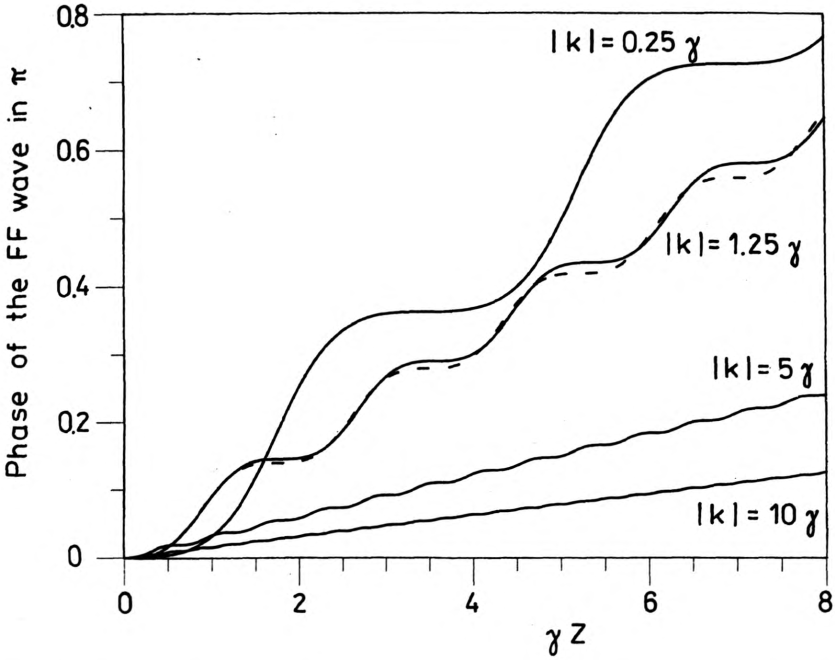

Fig. 1. Phase shift of the FF wave in the scalar ($\delta = 0$) interaction for different ratios mismatch/nonlinearity as a function of the normalised length $\xi = \gamma Z$ ($\gamma = \text{const}$). Solid line — exact results, dashed line — approximation (7) used

$|k|/\gamma = 1.25$. If $|k|/\gamma \gtrsim 2$, the typical differences between the phase shifts, provided by both methods, are even at $\xi \approx 6$ less than 10^{-3}. We note again that this approximation has to be handled with care if the nonstationary case is concerned. There the condition $|k|/\gamma \gtrsim 1$ means that the mismatch has to exceed the maximum nonlinearity set by the peak power of the pulse. The dynamics of the scalar (or type I) case (the only wave at FF interacts with one SH wave) can also be inferred from the analysis performed provided that the limit of the balanced input ($\delta = 0$) is taken. We mention that the diversity of the scalar case is much less than that identified for the vectorial one. In particular, it is easy to conclude looking at (2)–(6) and known from many papers that no phase modulation appears if $k = 0$. Finally, the phase modulation is always weaker than that for the weaker FF wave in the vectorial case.

3. Effective cubic nonlinear coefficient of the cascaded quadratic process

The efficiency of the $\chi^{(2)}$ interaction can be estimated by the nonlinear phase shift of the FF wave evoked by the cascaded process of up- and down-conversion which in turn can be compared with that accumulated in a degenerated cubic process. It is convenient to use the differential phase shift defined in a cubic medium as $d\varphi^{(3)}/dz = (\omega)/c)n_2 I$, where n_2 is the nonlinear coefficient commonly used and I is the input intensity (bulk) or the guided power divided by the effective core area of the respective mode (waveguide). Frequently one argues that cascaded quadratic processes resemble the cubic one provided that the mismatch is large ($|k| \gg 1$). Taking as an example a stationary scalar interaction ($\delta = 0$) we are going to show that this condition is inaccurate and too rough and that there are essentially three levels of approximation, where again the relation between nonlinearity γ and mismatch k plays a key role.

3.1. Large mismatch $|k| \gg 1$ and $|k| \gg \gamma$

The crudest approximation to deal with that case consists in rigidly locking the SH wave to the FF one ($db/dZ \approx 0$) in (1). Then we get a constant effective nonlinear coefficient of the cascaded process proportional to the square of the second-order coefficient and the inverse of the mismatch

$$(n_2^{\text{eff}})_1 = \frac{\chi_{\text{eff}}^2}{(\omega/c)\Delta\beta}. \tag{8a}$$

3.2. Mismatch large compared to the nonlinearity $|k| \gg \gamma$

A more reliable approach is based on the phase evolution obtained from Eq. (1) $d\varphi/d\xi = (k/\gamma)\{-1+[1/(1-u(\xi))]\}$, where $\varphi(\xi)$ is the phase of the FF wave, $u(\xi)$ is intensity of the SH, and $\xi = \gamma Z$. Expanding this expression into a Taylor series to the first order in $u(\xi)$ we get the relation $d\varphi/d\xi = (\gamma/2k)\{1-\cos[(2k/\gamma)\xi]\}$. Using the normalisations we can read off the effective nonlinear coefficient as

$$(n_2^{\text{eff}})_2 = \frac{\chi_{\text{eff}}^2}{(\omega/c)\Delta\beta}[1-\cos(\Delta\beta z)], \tag{8b}$$

which depends now on the propagation distance. This expression was earlier derived in the so-called non-depleted pump approximation [7]. Note that Eq. (8b) simplifies to (8a) if the trigonometric function varies rapidly ($|k| \gg 1$) and its average vanishes.

3.3. Moderate mismatch $|k| \gtrsim \gamma$

Finally we lift the strong constraint ($|k| \gg \gamma$) coming to the case $|k| \gtrsim \gamma$. The approximation used there corresponds to the replacement of the solution of (1) presented by elliptic functions by trigonometric ones [12] but avoids the Taylor expansion of $[1/1-u(\xi)]$. The straightforward calculation leads to

$$(n_2^{\text{eff}})_3 = \frac{\chi_{\text{eff}}^2}{(\omega/c)\Delta\beta}\,\frac{[1-\cos(\Delta\beta z)]}{[1-(4\chi_{\text{eff}}^2 P/\Delta\beta^2)\sin^2(\Delta\beta z/2)]} \tag{8c}$$

revealing that the effective nonlinear coefficient depends now on the propagation distance as well as on the input power (intensity in the bulk case). The factor in front of the sin-function corresponds to $(\gamma/k)^2$. Hence, it is evident that (8b) can be re-established if $(\gamma/k)^2 \ll 1$ holds. We can conclude that it might be useful to exploit the concept of an effective third-order nonlinear coefficient provided that, at least, the linear mismatch exceeds the nonlinearity $(2\chi_{\text{eff}}\sqrt{P}/\Delta\beta) \lesssim 1$. As already pointed

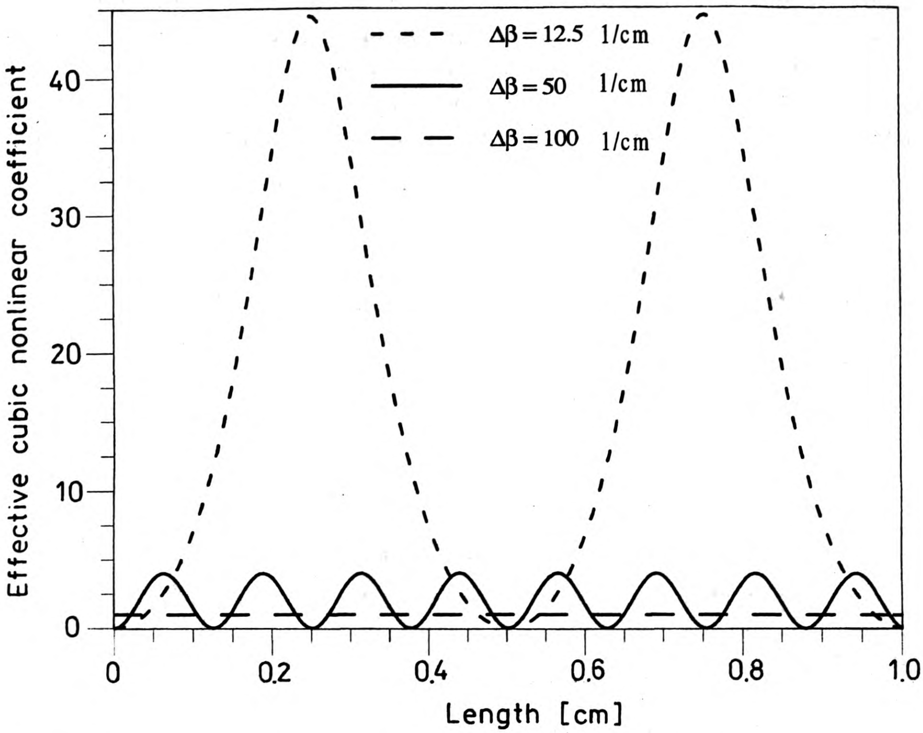

Fig. 2. Effective cubic nonlinear coefficient of the cascaded second-order process versus propagation length. Parameters: input irradiance 500 MW/cm^2, $d_{eff} = 3.1$ pm/V, $n = 1.8$, $\lambda = 1.06$ µm

out the crucial parameter which controls the characteristic of the cascaded process is the relation between mismatch k and nonlinearity γ rather than one of these quantities. The effective n_2 coefficient obtained in different approximations is plotted in Fig. 2 as a function of propagation distance for different values of phase mismatch $\Delta\beta$ in a KTP crystal. The pump irradiance is supposed to be 500 MW/cm^2 at 1.06 µm. Eventually, it remains to mention that second-order cascaded processes own a much richer diversity than those described by an effective cubic nonlinearity.

4. Amplitude and phase modulation of short pulses

Now it is commonly believed that cascading of quadratic nonlinearities could represent a competitive alternative to the conventional schemes based on degenerated cubic effects in future photonic networks [12] – [16]. Having this aspect in mind, it is necessary to study the amplitude and phase modulation for pulses in the picosecond regime. We mention that dispersion-evoked pulse broadening and temporal walk-off can be neglected in the context we focus on. Hence, it remains to evaluate the analytical formulas obtained in the previous sections to study the intensity dependence of the output modulation. First we recall that in a cubic material self-phase modulation due to the variation of the power across the pulse is the only effect appearing. Thus, it is unavoidable in these materials that the pulse acquires a strong chirp upon propagation. This is expected to be detrimental for all-optical switching, *e.g.*, in a Mach–Zehnder interferometer where a uniform phase shift of the entire pulse is required to obtain an effective amplitude modulation due to constructive or destructive interference.

As far as cascading is concerned this behaviour appears only in the limit of moderate or large mismatch, where the phase shift depends almost linearly on power and the amplitude modulation is only minor (Fig. 1). But for small or zero mismatch the coexistence of strong amplitude and phase modulation, evoked by the pronounced conversion process, is peculiar for cascading. The strong amplitude modulation depends on the normalised distance $\xi = \gamma Z$ which contains the power via the nonlinearity γ. Another peculiarity of the cascading process consists in the existence of pronounced plateaus, where the phase is almost constant provided that the control parameters are properly fixed (Fig. 1). We are going to discuss the consequences for phase modulation and assume that the complete initial pulse amplitude varies as $\sim\sqrt{P_0}\exp(-\tau^2)$, where τ is an arbitrarily normalised time in the reference frame of the pulse.

Inspecting Equations (5) and (6), one can show that essentially two cases of phase modulation can be distinguished, namely for small mismatch a monotonous, but almost step-like increase of the phase of the weaker FF wave and for moderate mismatch a quasi-linear increase of the phase with slight oscillations imposed (see Fig. 1).

By exploiting Equations (2)–(5), we can calculate the amplitude and phase modulation as a function of the nonlinearity which now varies across the pulse as $\gamma = \gamma_0\exp(-\tau)^2$ with $\gamma_0 = L\chi_{\text{eff}}\sqrt{P_0}$. For the moderate mismatch the approxima-

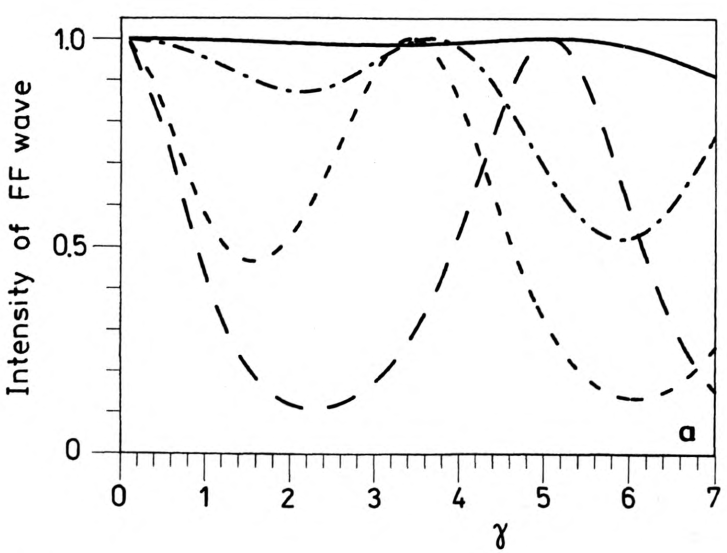

Fig. 3a

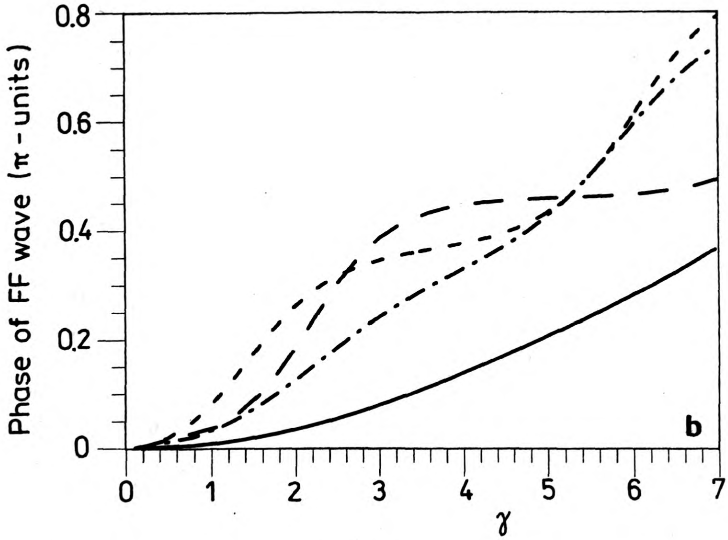

Fig. 3. Intensities (a) and phases (b) of the FF wave as a function of the nonlinearity $\gamma = L\chi_{\text{eff}}\sqrt{P}$. The input is balanced ($\delta = 0$); solid line: $k = 17.3$, dash-dotted line: $k = 4.3$, short-dashed line: $k = 0.87$, long-dashed line: $k = 0.25$

tion (7) can be used as well. The results are shown in Fig. 3 for various values of mismatch. Figure 3 reveals that for moderate mismatches the phase shift is proportional to γ^2 and depends thus linearly on the instantaneous intensity. Hence, we can expect a similar phase modulation as in cubic materials because the amplitude modulation is fairly weak.

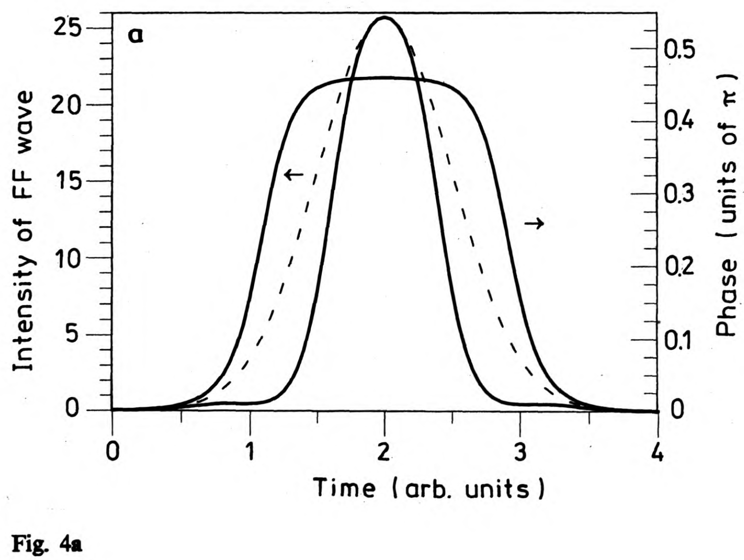

Fig. 4a

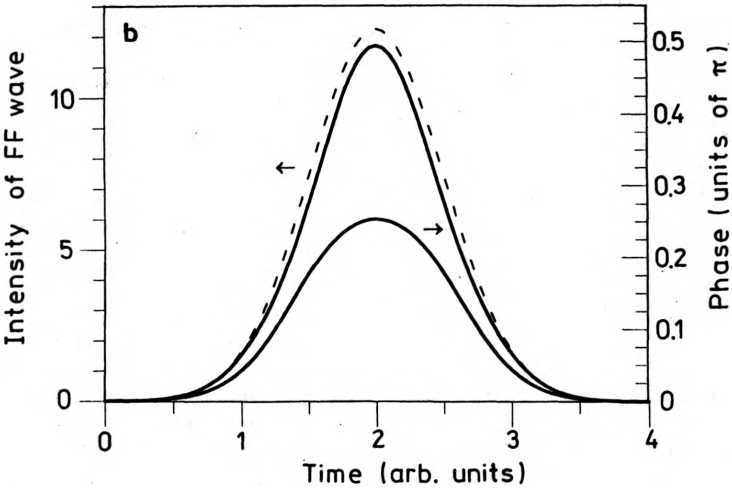

Fig. 4. Phase and amplitude modulation of a pulse with the input amplitude $\gamma = \gamma_0 \exp(-\tau^2)$. **a** – $k = 0.25$, $\gamma_0 = 5.07$, **b** – $k = 4.3$, $\gamma_0 = 3.5$

The consequences of these results for the intensities and the phases of the pulse can be recognised from Fig. 4. In Figure 4**a**, the peak nonlinearity was set to $\gamma_0 =$ 5.07 corresponding to a maximum amplitude of the FF waves and a centre of a phase plateau (see Fig. 3). For the cases where the mismatch is small (Fig. 4**a**), the pulse shapes are only marginally affected, whereas the phase modulation is almost homogeneous (no chirp) across the pulse. In contrast Fig. 4**b** shows the behaviour similar to that encountered in cubic materials. The phase modulation follows the input intensity of the pulse corresponding to a strong chirp.

5. Conclusions

In conclusion, we have shown that with regard to ultrafast all-optical switching cascading of quadratic nonlinearities can be qualitatively superior to the cubic effects. First, one can achieve a highly efficient direct intensity modulation at the output. Secondly, if phase modulation effects are exploited, being usually the arena of cubic nonlinearities, situations can be identified where the pulse shift is homogeneous across the entire pulse. Hence, devices which transform this phase shift into an intensity modulation as Mach–Zehnder interferometers, loop mirrors or phase rotators combined with crossed analysers are expected to operate much more efficiently if cascaded quadratic nonlinearities are exploited rather than cubic ones.

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