

# ON EFFICIENT FIELD ENERGY CONVERSION IN NON-PHASE-MATCHED FREQUENCY DOUBLING PROCESS

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In the paper the efficiency of the light energy conversion from the fundamental wave into its second harmonics is analysed. A standard model of three waves mixing in media without centre of symmetry was applied. It was shown that the full conversion of energy is possible if an appropriate phase difference exists and provided that at least a minimal energy of the second wave is present on the input. The results are expressed both by analytical formulas and a phase space reconstruction. A simple experimental setup to enhance the second harmonics is proposed.

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## 1. Introduction

The possibility of energy transfer between light waves of different frequencies is of great interest in modern nonlinear optics [1]. Especially important is the process of second harmonics generation (SHG) which appears in media without centre of symmetry (exhibiting the second-order nonlinearity). The problem of making this process more effective is the principal one [2] as it is strictly related to the action of radiation converters. In particular, second harmonics generation might be an efficient source of the blue light laser beams. It is commonly accepted [1-3] that such a process is not possible under finite value of the phase-mismatch parameter. It is practically difficult to ensure full synchronisation of phases of two beams that are spatially bound and that this condition is crucial in the conversion process. There exist several methods of enlarging the efficiency of the conversion; some of them are described in [3]. They are based mostly on the natural anisotropy of the refraction index in media exhibiting dichroism. In the case when the anisotropy exceeds the dispersion effect between first and second wave, there exists an angle

of full phase matching. The electric field vector  $\mathbf{E}_\omega$  oscillates perpendicularly to the plane determined by the main axis of the crystal and wave vectors  $\mathbf{k}_\omega$  and  $\mathbf{k}_{2\omega}$ , vector  $\mathbf{E}_{2\omega}$  lies then in this plane being perpendicular to both wave vectors. In this configuration, the phase-mismatch parameter  $\Delta k = |\mathbf{k}_\omega - \mathbf{k}_{2\omega}|$  vanishes, thus full phase synchronisation appears. Full conversion is however impossible because Poynting vectors of both beams are not collinear. The beams diverge, therefore the region of interaction of beams is limited. Besides spatial dimensions of them are small [4]. These factors stand in contradiction with the condition  $\Delta k = 0$ .

In this paper we suggest that the condition  $\Delta k = 0$  is, in general, not necessary for the full conversion. Instead of that we should ensure a non-zero component of doubled frequency at the input. Additionally, its phase must be properly matched with regard to the phase of the fundamental beam. We derive analytical formulas for this minimal amount of the second harmonics, necessary for the full energy conversion. As a special case we obtain from them formulas corresponding to the standard condition  $\Delta k = 0$ . We present also adequate phase portraits of the mixing process for several values of the phase-mismatch parameter  $\Delta k$ . As a conclusion we suggest a simple experimental model allowing effective doubling of the laser beam.

## 2. Quantitative analysis of the second harmonics generation

We consider two beams with frequencies  $\omega$  and  $2\omega$  incident perpendicularly on a transparent medium without centre of symmetry. Let the main optical axis be parallel to wave vector  $\mathbf{k}$ . According to this assumption the length of interaction is sufficiently large. This direction will be called  $Oz$  axis. The nonlinear properties of the medium are described by the third-rank tensor  $\chi^2(\omega)$ . The equations describing the evolution of  $\mathbf{E}$  for both waves in the medium have a standard form [1–3]:

$$\frac{d}{dz} E_{2\omega} = i\kappa_{2\omega} E_\omega^2 e^{-i\Delta k z}, \quad (1)$$

$$\frac{d}{dz} E_\omega = i\kappa_\omega E_{2\omega} E_\omega^* e^{-i\Delta k z},$$

where the coupling constants  $\kappa_p$  are defined as

$$\kappa_p = \frac{p}{2n_p c} \chi^{(2)}(p) \quad (p = \omega, 2\omega), \quad (2)$$

and  $n_p$  are linear refraction coefficients for both frequencies. The mismatch parameter  $\Delta k$  is given by the formula

$$\Delta k = \frac{2\omega}{c} (n_\omega - n_{2\omega}). \quad (3)$$

In this paper the following normalisation is used for the electric field:

$$a_p = \sqrt{\kappa_\omega \kappa_p} L |E_p|, \quad \varphi_p = \arg(E_p). \quad (4)$$

Moreover, we normalise the coordinate  $z$  by introducing the new variable  $s = z/L$ . Similarly, the new mismatch parameter is equal  $\Delta K = \Delta k L$ . These transformations allow to write Eqs. (4) in the form

$$\frac{d}{ds} a_{2\omega} = a_\omega^2 \sin \theta,$$

$$\frac{d}{ds}a_\omega = -a_\omega a_{2\omega} \sin \theta, \quad (5)$$

where the phase variable  $\theta = \varphi_{2\omega} - 2\varphi_\omega + \Delta K s$  fulfils the equation

$$\frac{d}{ds}\theta = \left( \frac{a_\omega^2}{a_{2\omega}} - 2a_{2\omega} \right) \cos \theta + \Delta K. \quad (6)$$

The general solution of the two-wave mixing process is a special case of the solution for the non-degenerate three wave mixing processes given in [5]. In this case one obtains expressions in the form of elliptic functions of Jacobi and of Weierstrass. If  $\Delta k = 0$  and if the second harmonics is absent on the input ( $u_{20} \equiv u_{2\omega}(0) \equiv a_{2\omega}^2(0) = 0$  but  $u_{10} \equiv u_\omega(0) \equiv a_{2\omega}^2(0)$ ) one gets the well known formulas [1-3]:

$$a_{2\omega}^2(s) \equiv u_{2\omega}(s) = u_{10} \tanh^2 \sqrt{u_{10}} s, \quad (7)$$

$$a_\omega^2(s) \equiv u_\omega(s) = u_{10} \operatorname{sech}^2 \sqrt{u_{10}} s,$$

corresponding to the full conversion of basic beam. In a more general case when  $u_{20} \neq 0$  (but still  $\Delta k = 0$ ) for  $\theta_0 \equiv \theta(s=0) = \pm\pi/2$  one may derive (see [5]) the expression

$$\begin{aligned} u_{2\omega}(s) &= (u_{10} + u_{20}) \tanh^2 \left( \tanh^{-1} \sqrt{\frac{u_{20}}{u_{10} + u_{20}}} + \sqrt{u_{10} + u_{20}} s \right), \\ u_\omega(s) &= (u_{10} + u_{20}) \operatorname{sech}^2 \left( \tanh^{-1} \sqrt{\frac{u_{20}}{u_{10} + u_{20}}} + \sqrt{u_{10} + u_{20}} s \right). \end{aligned} \quad (8)$$

Hence we have a possibility of the full conversion even for  $\Delta k = 0$ , provided the appropriate relation between phases of incident beams.

If  $\Delta k \neq 0$ , then for  $u_{20} \geq \Delta K^2/4$  and

$$\cos \theta_0 = \frac{|\Delta K|}{2\sqrt{u_{20}}}, \quad (9)$$

the general solution has the form [5]:

$$\begin{aligned} \tilde{u}_{2\omega}(s) &= (u_{10} + \tilde{u}_{20}) \tanh^2 \left( \tanh^{-1} \sqrt{\frac{\tilde{u}_{20}}{u_{10} + \tilde{u}_{20}}} + \sqrt{u_{10} + \tilde{u}_{20}} s \right), \\ u_\omega(s) &= (u_{10} + \tilde{u}_{20}) \operatorname{sech}^2 \left( \tanh^{-1} \sqrt{\frac{\tilde{u}_{20}}{u_{10} + \tilde{u}_{20}}} + \sqrt{u_{10} + \tilde{u}_{20}} s \right), \end{aligned} \quad (10)$$

where we have used the abbreviation

$$\tilde{u}_{2\omega} = u_{2\omega} - \frac{\Delta K^2}{4}. \quad (11)$$

In the extreme case  $u_{20} = \Delta K^2/4$ , we obtain simpler expressions for the normalised amplitudes

$$\tilde{u}_{2\omega}(s) = u_{10} \tanh^2 \sqrt{u_{10}} s, \quad (12)$$

$$u_\omega(s) = u_{10} \operatorname{sech}^2 \sqrt{u_{10}} s.$$

It is easy to see that formulas (10) and (12) are analytically identical with expressions (7) and (8) which were obtained for the SHG process with  $\Delta k = 0$ .

### 3. Qualitative analysis of the SHG process

A possibility of the full conversion for  $\Delta k \neq 0$  was concluded from exact solutions of Eq. (1). One can also analyse the wave mixing processes by means of methods used in the theory of differential equations [6]. Qualitative features of dynamic systems may be deduced without solving the equations. It is sufficient to analyse the topology of trajectories of the system in the generalised "phase space" (not to be mistaken with the phase of a wave).

In order to construct such a space we introduce a new variable (the amplitude parameter):

$$\zeta = \frac{u_{2\omega}}{u_{\omega} + u_{2\omega}}. \quad (13)$$

It should be stressed that, for a lossless medium, the denominator of the above expression is constant.

It is easy to verify that

$$(1 - \zeta)\sqrt{\zeta} \cos \theta + \sqrt{\delta} \zeta = \text{const} \equiv C. \quad (14)$$

By introducing a new parameter

$$\delta = \frac{\Delta K^2/4}{u_{\omega} + u_{2\omega}} \quad (15)$$

we obtain an intensity-dependent representation of the phase-mismatch.

The variable  $\zeta$  fulfils the condition

$$0 \leq \zeta \leq 1. \quad (16)$$

Thus, for a given  $\delta$ , the function  $C(\zeta, \theta)$  determines a surface. The most interesting are the lines of constant value of  $C$ , hereafter called "trajectories", because they correspond to real dynamics of the system for which Eq. (14) holds.

In Fig. 1 we have shown trajectories (on a phase portrait) for different values of  $\delta$ . Bold lines depict two extreme trajectories. First of them (full line) corresponds to the full conversion of radiation, for which  $\zeta = 1$ . As

$$\lim_{\zeta \rightarrow 1} C(\zeta, \theta) = \sqrt{\delta}, \quad (17)$$

this limit trajectory represents the trajectory  $C = \sqrt{\delta}$ . Another limit trajectory (dotted line) corresponds to the zero value of the second harmonics at the input ( $\zeta_0 = 0$ ), thus we have now to do with the strict SHG process. The formula (14) implies that  $C = 0$ . From this figure it is easy to see that for  $\delta = 0$  (hence  $\Delta K = 0$ ) the full conversion appears only when  $\zeta_0 = 0$  (no second harmonics on the input). The angle corresponding to these trajectories  $\theta = \pm\pi/2$ . On the other hand, if  $\zeta_0 < 1/3$ , then the conversion between two waves has oscillatory character. The efficiency of this conversion gradually decreases and for  $\zeta_0 = 1/3$  reaches zero value. In this case intensities of both two waves remain constant through the whole interaction length. This corresponds to the notion of eigenmodes derived in [7]. From Eq. (14) it also follows that such a situation exists for all values of  $u_1$  i  $u_2$ , for which the boundary condition  $u_{20}/u_{10} = 1/2$  is fulfilled.

Figures 1a and b are qualitatively similar. However, the shape of the trajectories  $C = \sqrt{\delta}$  is different in both cases. Figure 1b indicates the possibility of full conversion for  $\zeta_0 \geq \delta$  (or  $u_{20} \geq \Delta K^2/4$ ). This process is restricted by (15), hence for  $\delta \geq 1$  the full conversion is not possible, which is in agreement with Fig. 1c.

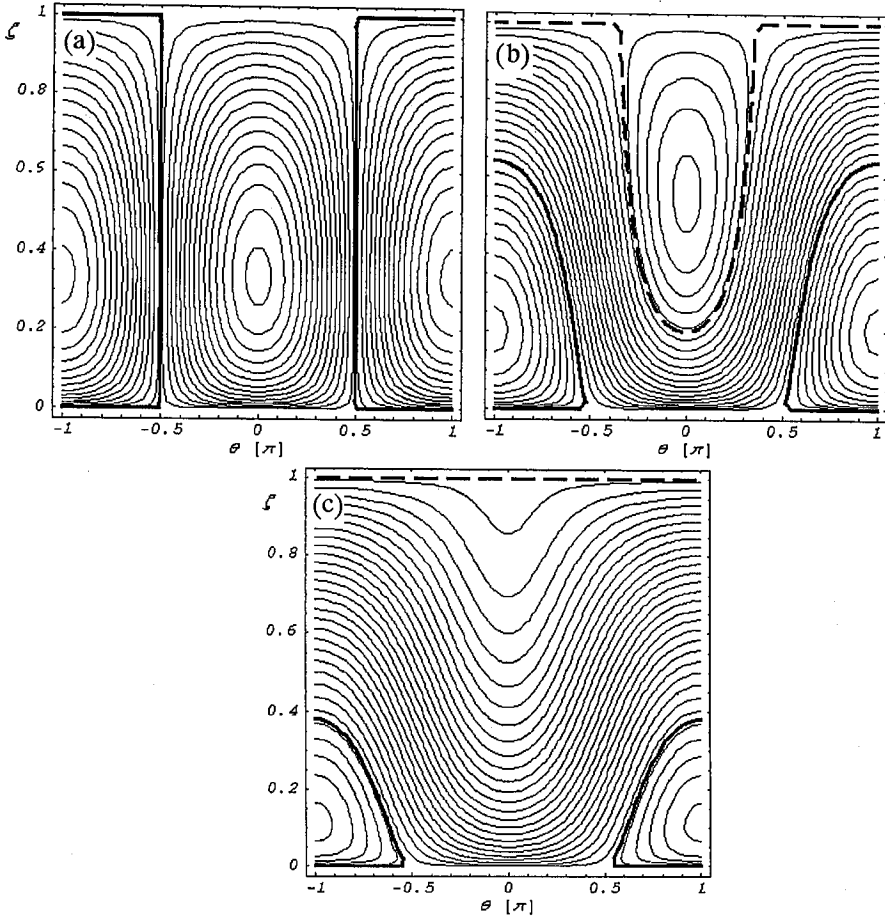


Fig. 1. Phase portraits for three values of the conversion parameter  $\delta$ : (a)  $\delta = 0$ , (b)  $\delta = 0.2$ , (c)  $\delta = 1$ . The solid line corresponds to the trajectory for which  $\zeta(s = 0) = 0$ ; the dotted line is the limit trajectory  $\zeta(s) \rightarrow 1$ , for which one can obtain the full conversion of a monochromatic wave into its second harmonics.

#### 4. Optical SH generator

Phase portraits method suggests how to design a simple experimental setup for conversion of a wave into its second harmonics in the lack of phase synchronism. A scheme of such converter is shown in Fig. 2. It consists of two appropriate plates made of nonlinear transparent material of second-order nonlinearity; a slab of a linear transparent material (for instance glass) fills the space between them. In the first plate some amount of second harmonics is generated. Intermittent material shifts the phase of this wave with regard to the basic wave; its thickness is chosen in such a way that the system achieves the trajectory which enables the full conversion. It may be realised in the next plate. Instead of matching the thickness we may appropriately choose the intensity of the incident beam.

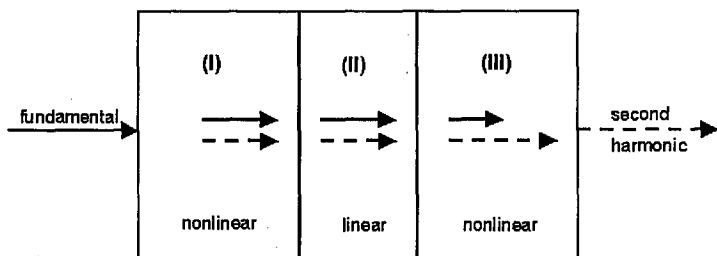


Fig. 2. A scheme of an optical frequency converter: in the plate I (made of a nonlinear material) an amount of second harmonics is generated; the second plate made of linear material appropriately shifts its phase. On the output from the plate III (made of nonlinear material) one gets a beam consisting of the second harmonics only.

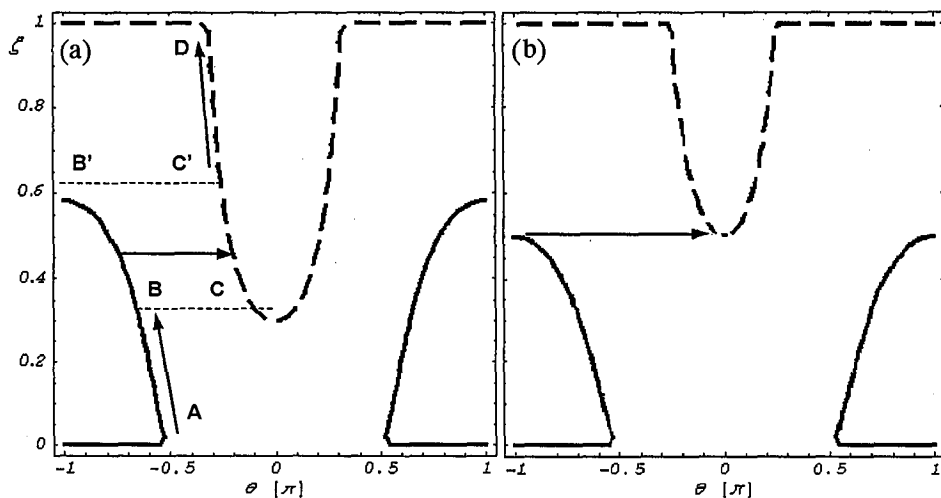


Fig. 3. Phase portraits illustrating the principle of action of an optical converter. The solid line shows the evolution of the second harmonics in the first plate whereas the dotted line — the evolution in the third plate. (a)  $\delta = 0.3$ ; the intensity of the second harmonics evolves along the line  $A \rightarrow B(B') \rightarrow C(C') \rightarrow D$ ; (b) the limit case  $\delta = 0.5$ .

The possibility of full conversion is restricted to cases for which  $\delta \leq 1/2$ , or  $\Delta K^2 \leq 2u_\omega(s=0)$  (see Fig. 3a). The extreme case  $\delta = 1/2$  is illustrated in Fig. 3b. If

$$\frac{1}{2} < \delta < 1 \quad (18)$$

(or  $2u_{10} < \Delta K^2 < 4u_{10}$ ), it is possible to apply a set of plates (a sandwich structure).

Another possibility of steering the phase shifts in linear slabs might be based on the Pockels (or Kerr) effects. Refraction index depends on the applied field intensity (or its square). By changing the value of external field one may get the desired value of phase difference between two waves.

## 5. Summary

We have presented here a model of efficient conversion of a monochromatic wave into its second harmonics in the absence of phase synchronism. The results of calculations, as well as phase portraits analysis, lead to the conclusion that it is possible to obtain the full conversion and show the way how to construct a frequency converter. We propose a simple setup that can be used to check this idea. Besides, our results enlarge the class of interesting non-centrosymmetric materials by these in which the optical dispersion is essentially smaller than the optical anisotropy. It implies also smaller absorption [4] that is important in signal processing and transmitting.

It is also worthwhile to mention the advantages of the technique based on phase portraits. Due to this one can obtain interesting (both qualitative and quantitative) results without solving complicated equations of motion.

## References

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