

Nonlinear phase change in type II second-harmonic generation under exact phase-matched conditions

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The process of type II second-harmonic generation has been investigated theoretically. It is found that, unlike in the process of type I second-harmonic generation, phase matching is not necessary for a significant shift of the pump wave phase to be produced. The simple inequality of the intensities of interacting waves suffices for this effect to occur.

Late in the 1960's the strong self-action of pump waves by means of the reaction of a second-harmonic wave in the process of second-harmonic generation (SHG) was predicted.¹ Recently interest in cascaded second-order nonlinearity has been rekindled because of the prospects of using this nonlinearity in all-optical devices, e.g., nonlinear Mach-Zehnder interferometers, nonlinear direction couplers,² and optical transistors.³

The effect of the nonlinear phase change of a pump wave caused by cascaded second-order nonlinearity has been observed in KTiOPO_4 crystal.⁴ In Refs. 1 and 4 the theory of the nonlinear phase change in the type I SHG process has been developed, and it has been shown that there is no phase change in the type I SHG process under exact phase matching, $\Delta\mathbf{k} = \mathbf{k}(2\omega) - 2\mathbf{k}(\omega) = 0$. The effect appears only when the wave-vector mismatch is nonzero. The situation is quite different for the type II SHG process, and this is shown below.

In this Letter we consider the type II SHG process and the conditions when the nonlinear phase shift is possible.

The type II SHG process is characterized by the interaction of two pump waves that have different polarizations. Thus this interaction is nondegenerate, and the conditions for phase matching are

$$\begin{aligned} \mathbf{k}_1(\omega) + \mathbf{k}_2(\omega) &= \mathbf{k}_3(2\omega), \\ \mathbf{k}_1(\omega) \neq \mathbf{k}_2(\omega) &\neq \frac{1}{2}\mathbf{k}_3(2\omega). \end{aligned} \quad (1)$$

The nondegenerate three-wave interaction in the approximation of a slowly varying amplitude is given by the following system of equations:

$$\begin{aligned} \frac{\partial E_1}{\partial z} &= \frac{i\omega}{2n_1c} \chi^{(2)}(\omega; 2\omega, -\omega) E_2^* E_3 \exp(-i\Delta kz), \\ \frac{\partial E_2}{\partial z} &= \frac{i\omega}{2n_2c} \chi^{(2)}(\omega; 2\omega, -\omega) E_1^* E_3 \exp(-i\Delta kz), \\ \frac{\partial E_3}{\partial z} &= \frac{i2\omega}{2n_3c} \chi^{(2)}(2\omega; \omega, -\omega) E_1 E_2 \exp(i\Delta kz), \end{aligned} \quad (2)$$

where E_i are the complex amplitudes of the electric fields of the interacting waves, n_i are the refractive

indices, $\Delta k = k_1(\omega) + k_2(\omega) - k_3(2\omega)$ is a wave-vector mismatch, the subscripts $i = 1, 2$ correspond to pump waves 1 and 2, and the subscript $i = 3$ corresponds to the wave of the second harmonic.

Below we consider only media without any dissipation, and hence Kleinman symmetry relations can be applied and Eqs. (2) can be expressed in terms of common nonlinear d coefficients by $d_{\text{eff}} = |\chi^{(2)}(2\omega; \omega, \omega)|/2$. Introducing the scaling of the amplitudes as

$$A_i = E_i \left(\frac{n_i}{2I_0} \sqrt{\frac{\varepsilon_0}{\mu_0}} \right)^{1/2},$$

where ε_0 and μ_0 are the dielectric and magnetic permeabilities of the vacuum and I_0 is the normalizing intensity, we are led to the following system:

$$\begin{aligned} \frac{\partial A_1}{\partial z} &= i\Gamma A_2^* A_3 \exp(-i\Delta kz), \\ \frac{\partial A_2}{\partial z} &= i\Gamma A_1^* A_3 \exp(-i\Delta kz), \\ \frac{\partial A_3}{\partial z} &= i2\Gamma A_1 A_2 \exp(i\Delta kz), \end{aligned} \quad (3)$$

where

$$\Gamma = \frac{\omega}{c} \frac{d_{\text{eff}}}{\sqrt{n_1 n_2 n_3}} \left(2I_0 \sqrt{\frac{\mu_0}{\varepsilon_0}} \right)^{1/2}.$$

The solution of system (3) was carried out numerically. An analysis of the results is presented below. The calculated value of Γ was taken to be equal to 15 cm^{-1} , which corresponds to the normalizing power $I_0 = 1 \text{ GW/cm}^2$ and the material constants of KTiOPO_4 (Ref. 5) for pump waves of $1.064\text{-}\mu\text{m}$ wavelength ($n_1 = 1.8297$, $n_2 = 1.7421$, and $n_3 = 1.7859$ and $d_{\text{eff}} = 7.33 \times 10^{-12} \text{ m/V}$ for the propagation direction with respect to the main crystal axes $\theta = 90^\circ$ and $\varphi = 23.7^\circ$).

Let us consider the case for which the conditions of phase matching are exactly satisfied, i.e., $\Delta k = 0$. The results of the numerical calculations of the power and the phase evolutions with distance for this case are presented in Figs. 1 and 2. If the powers of the pump waves are equal, these dependencies are the

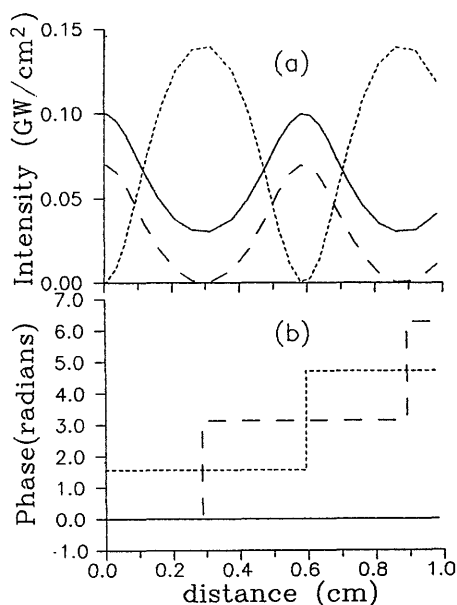


Fig. 1. Evolution of (a) the intensities and (b) the phases of two pump waves (dashed curves, wave 1; solid curve, wave 2) and the second-harmonic wave (dotted curves) for exact phase matching. The input intensities of the pump waves are unequal ($P_1 = 0.07 \text{ GW/cm}^2$, $P_2 = 0.1 \text{ GW/cm}^2$).

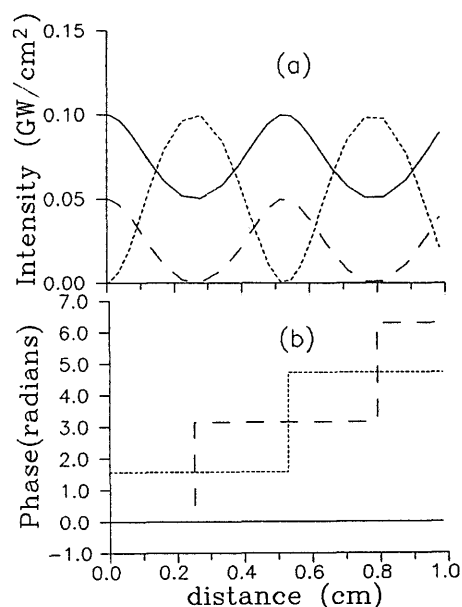


Fig. 2. Evolution of (a) the intensities and (b) the phases of two pump waves (dashed curves, wave 1; solid curve, wave 2) and the second-harmonic wave (dotted curves) for exact phase matching. The input intensities of the pump waves are unequal ($P_1 = 0.05 \text{ GW/cm}^2$, $P_2 = 0.1 \text{ GW/cm}^2$).

same as for type I phase matching.⁶ The phases of the interacting waves do not change with distance; they have a relationship such that the energy conversion goes into the second harmonic but does not go in the opposite direction. The nonperiodicity of this process may be understood under more careful consideration of the system of Eqs. (3). Indeed, the driving force [the right-hand sides of Eqs. (3)] at the

frequency 2ω produces a second-harmonic wave with such a phase that the driving force at the frequency ω is in opposite phase to the pump waves. In addition, the Manley-Rowe relations would hold for nondissipative media [see, for example, Eq. (2.50) of Ref. 6], which in our case are

$$\Delta|A_1|^2 = \Delta|A_2|^2 = -\frac{1}{2} \Delta|A_3|^2,$$

where $\Delta|A_i|^2$ is the change of intensity of the i th wave. It is evident that, if the pump intensities at the input are equal, the intensities are exhausted simultaneously. When this occurs all driving forces become equal to zero, and the process become irreversible.

An essentially different picture is observed if the intensity of one pump wave at the input to the nonlinear medium is unequal to the other. Figure 1 shows the situation in which the initial intensities differ by 1.4 times. One can see that the process of the energy exchange between the interacting waves is periodic under these conditions. The power of the pump wave with smaller initial value goes to zero at regularly spaced intervals, and the power of the other pump wave remains nonzero. At the same time the phase of the first wave suffers a steplike change of π . At the same moment the direction of the process is changed to the side of downconversion and remains the same until the power of the second-harmonic wave becomes zero. At that moment the phase of the second harmonic is changed by π , and the process changes direction to the side of upconversion. Then the process is repeated.

To clarify the physical sense we return to the system of Eqs. (3). Let the power of pump wave 1 be smaller than that of pump wave 2. It is clear from the Manley-Rowe relations that in this case the power of wave 2 is nonzero when the power of wave 1 is zero. As this takes place, the driving forces in the second and third equations of Eqs. (3) are zero, and the nonzero driving force produces a wave with frequency ω polarized as the initial wave 1 and having its phase shifted by π with respect to that of the initial wave 1. The phase change leads to a change of the process direction to that of the downconversion. The intensity of the second harmonic starts to be diminished; at the moment of its complete depletion the driving forces in the two first equations of Eqs. (3) become zero. The nonzero force in the third equation produces a wave with the frequency 2ω with its phase shifted with respect to the phase of the second harmonic before it becomes zero. The phase change of the second-harmonic wave leads to a change of the process direction to that of the side of upconversion.

Figure 2 shows the evolution of the energy-exchange period with a change in the ratio of the powers of the pump waves at the input to the nonlinear medium. Figure 2 shows the reduction of this period with decreasing power of the smaller wave while the power of the other pump wave remains constant.

The process of type II SHG under nonzero wave-vector mismatch and unequal pump-wave powers is shown in Fig. 3 and has the following characteristics:

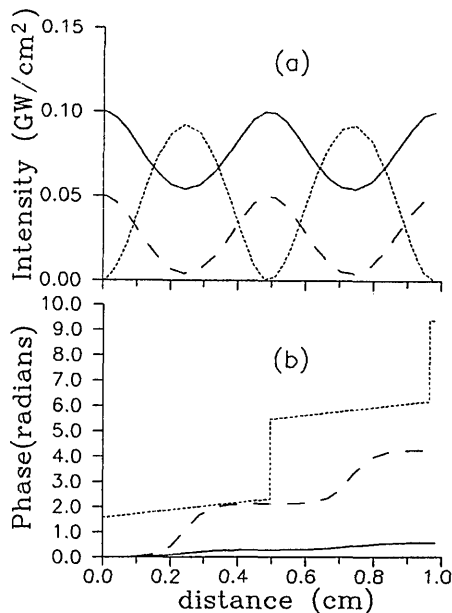


Fig. 3. Evolution of (a) the intensities and (b) the phases of two pump waves (dashed curves, wave 1; solid curves, wave 2) and the second-harmonic wave (dotted curves) for the case of nonzero wave-vector mismatch, $\Delta k = 3 \text{ cm}^{-1}$. The input intensities of the pump waves are unequal ($P_1 = 0.05 \text{ GW/cm}^2$, $P_2 = 0.1 \text{ GW/cm}^2$).

(1) The amplitude of the pump wave with smaller input power never becomes zero.

(2) The nonlinear phase change of this wave in one period is less than π and is reduced with the growth of the mismatch.

(3) The phase change of the pump wave with larger power is also nonzero.

(4) The amplitude of the second-harmonic wave periodically becomes zero, and its phase at these moments shifts by π .

(5) The period of energy exchange decreases with the growth of the wave-vector mismatch.

Under equal powers of the pump waves in type II SHG and with nonzero wave-vector mismatch, the characteristics of the process are equal to those of the one described in Ref. 4.

In conclusion, the remarkable peculiarities of the type II SHG process have been clarified. It has been determined that the process is periodic under exact phase matching and unequal initial intensities of the pump waves. The most remarkable point is that under the same conditions the phase of the wave with a smaller intensity exhibits an abrupt (steplike) change of π .

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