Nonlinear phase shift without cascaded second-order processes and third order nonlinearity

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Abstract

The new mechanism for obtaining a nonlinear phase shift has been proposed and the schemes are described for its implementation. As it is shown, the interference of two waves with intensity-dependent amplitude ratio coming from the second harmonic generation should produce the nonlinear phase shift. The sign and amount of nonlinear distortion of a beam wavefront is dependent of the relative phase of the waves that is introduced by the phase element. Calculated value of \( n_{\text{eff}}^2 \) exceeds that connected with cascaded quadratic nonlinearity, at the same conditions.

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There are a lot of potential applications for materials which exhibit a nonlinear phase shift (NPS) of the incident beam due to a local change of the refractive index. For instance, the nonlinear phase shift is necessary to obtain for the new generation of all-optical devices. Typically nonlinear phase shift has been achieved with an intensity-dependent refractive index coefficient \( n_2 \) \( (n = n_0 + n_2 I) \) due to third-order nonlinear susceptibility \( \chi^{(3)} \). Currently another nonlinear physical process that produces a nonlinear distortion in the phase of a beam appears to be of great interest \[1\] \[2\] \[3\] \[4\] \[5\] \[6\]. The phase shift utilizes the cascading of two nonlinear second-order processes: the up-conversion of a fundamental beam to a second harmonics (SH) and its subsequent downconversion back to the fundamental. A number of possible schemes of all-optical devices are discussed in Ref. \[6\].

This letter theoretically predicts a new mechanism for nonlinear phase shift. It is introduced when two waves with intensity-dependent amplitude ratio interfere. We also suggest optical schemes that one can use to obtain the nonlinear phase shift of both fundamental wave and its second harmonics.
The estimations show that the magnitude of the nonlinear shift may be several times as large as the one occurring due to cascaded processes under the same value of exciting intensity and nonlinear medium length.

The essence of the proposed technique can be clarified by the following simple considerations.

The phase \( \Phi \) of the wave produced by interference of two waves is given, as it is known, by the expression:

\[
\sin \Phi = \frac{\sin \phi_1 + \frac{a_2}{a_1} \sin \phi_2}{\sqrt{1 + \frac{a_2^2}{a_1^2} + 2 \frac{a_2}{a_1} \cos(\phi_1 - \phi_2)}}
\]

(1)

where \( a_1, a_2, \phi_1, \phi_2 \) are the corresponding amplitudes and phases of the interfering waves. If we happen to produce a dependence of the ratio \( a_2/a_1 \) upon intensity, obviously the phase \( \Phi \) becomes dependent of the intensity. To produce this intensity-dependent ratio \( a_2/a_1 \), we will use second harmonics generation.

Further calculations are shown on the example of two schemes. The first allows one to have NPS in the second-harmonics beam, and the other outputs an NPS in the fundamental wave.

The first scheme (fig. 1a) is essentially, a dispersion interferometer that makes use of orthogonally polarized second harmonic waves [7]. Both waves in the dispersion interferometer propagate along the same path. Waves of different frequencies correspond to the two arms of the interferometer, and the relative phase is gained owing to the dispersion of the phase element. In fig. 1a: 1,3 — frequency doublers that use type II phase matching (although an analogous scheme can be constructed for the type I), 2 — phase element (like an optical wedge with a compensator), 4 — filter that cuts of the fundamental wave, 5 — polarizer or polarizing beam-splitter. The elements have the following orientation: the frequency doublers are oriented orthogonally, the fundamental wave is polarized at 45° angle with respect to the optical axes of the crystals. The axis of the polarizer 5 is at the same angle as the polarization of the fundamental wave.

The described interferometer has following notable features owing to orthogonal polarization of the two second harmonic waves. First, the second harmonics generated by the first doubler is not downconverted inside the
second crystal due to the large phase mismatch. Second, the SH waves are generated in the first and the second crystals independently of each other and interfere behind polarizer 5. Let’s write the complex amplitudes of the two second harmonic waves as $a_1^{(2\omega)} \exp(i\phi_1)$, $a_2^{(2\omega)} \exp(i\phi_2)$. The relative phase ($\phi_2 - \phi_1$) can be adjusted with a phase element, in our case, due to variable thickness of the optical wedge $\Delta l$: 

$$\Delta(\phi_2 - \phi_1) = \frac{2\pi \Delta l}{\lambda/2} [n(\omega) - n(2\omega)]$$

where $n(\omega)$ and $n(2\omega)$ are refractive indices of the wedge material at frequencies $\omega$ and $2\omega$. Assume that the nonlinear phase effects inside each crystal are absent or negligible, i.e. $\phi_1$ and $\phi_2$ are independent of the intensity. This implies the following conditions:

$$\Delta k l_{1,2} \ll 1; \quad \frac{n_2 I_{l_{1,2}}}{\lambda} \ll 1,$$

where $l_1, l_2$ are the lengths of the first and second crystals, correspondly, $\Delta k$ — wave-vector mismatch between the fundamental and second harmonics, $I$ — the incoming fundamental intensity. The stated approximation essentially means that cascaded second-order processes and third-order nonlinearity do not result in any tangible phase shifts.

Let’s show that the phase of the resulting wave $\Phi$ at the exit from the dispersion interferometer depends on intensity. In the simplest case, when one can neglect the group velocity dispersion, beam walk-off effect and use exact phase matching approximation $\Delta k = 0$, amplitudes $a_1^{(2\omega)}$ and $a_2^{(2\omega)}$ are given by (3):

$$a_1^{(2\omega)} = I^{1/2} \text{th} \left( l_1 4\pi \chi^{(2)} I^{1/2} \right),$$

$$a_2^{(2\omega)} = \left[ I - (a_1^{(2\omega)})^2 \right]^{1/2} \text{th} \left( l_2 4\pi \chi^{(2)} \left[ I - (a_1^{(2\omega)})^2 \right]^{1/2} \right).$$

Expressions (3) holds true while there is no initial second harmonics in the incident light. In the second crystal this condition is maintained owing to orthogonal orientation of the crystals. Behind polarizer 5, the waves interfere having the amplitudes $a_1^{(2\omega)}/\sqrt{2}$ and $a_2^{(2\omega)}/\sqrt{2}$. Thus, the dependence of the amplitude ratio of the interfering waves can be calculated with the expression:

$$\frac{a_2^{(2\omega)}}{a_1^{(2\omega)}} = \frac{(1 - \text{th}^2 \mathcal{K} l_1 I^{1/2})^{1/2} \text{th} \left[ \mathcal{K} l_2 I^{1/2} (1 - \text{th}^2 \mathcal{K} l_1 I^{1/2}) \right]}{\text{th} \mathcal{K} l_1 I^{1/2}},$$

(4)
where $\mathcal{K} = 4\pi\chi^{(2)}$.

Let’s analyze qualitatively Eqs. (1), (4). For $I^{1/2} \leq \frac{1}{\mathcal{K}l_1}$ we have $\alpha \sim \alpha$ and, hence, can simplify the formula:

$$
\frac{a_2^{(2\omega)}}{a_1^{(2\omega)}} \approx \frac{l_2}{l_1} \left(1 - \mathcal{K}^2 l_1^2 I\right).
$$

If $l_2 \gg l_1$, then $a_2^{(2\omega)}/a_1^{(2\omega)} \gg 1$ at low intensity and falls down as the intensity grows. In accordance with (1), at low intensity $\Phi \approx \phi_2$, and at high intensity phase $\Phi$ is led to phase $\phi_1$.

The dependence of nonlinear phase shift $\Delta\Phi_{NL}$ upon dimensionless input intensity $\mathcal{K}^2 l^2 I$ (where $l = l_1 + l_2$) is shown in fig. 1b, calculated according to (1), (4). The maximal nonlinear phase shift possible to achieve is limited to the relative phase $\phi_2 - \phi_1$, that explains the saturation of the curve in fig. 1b.

In fig. 2, we see the dependence of $\Delta\Phi_{NL}$ and $I_{2\omega}/I$ upon the relative phase $(\phi_2 - \phi_1)$. The nonlinear phase shift can be either positive (leading to beam self-defocusing), or negative (self-focusing), depending on $(\phi_2 - \phi_1)$. It is easily seen that the maximum shift is produced at $\phi_2 - \phi_1 \approx \pi n$, however the SH output in contrast to it is at minimum, despite of nearly 100% second-harmonic generation efficiency at the given exciting intensity. The reason is that only a part of SH radiation passes through the polarizer, namely, the component which has the nonlinear phase shift. Of course, the phase of the fundamental wave in this scheme is not dependent of the intensity.

To obtain a nonlinear phase shift of the fundamental wave we can suggest as an example a scheme similar to a polarizing interferometer with the use of frequency doublers. The mechanism of the nonlinear phase shift here is analogous to that of the dispersion interferometer, namely, due to the interference of the waves with intensity-dependent amplitude ratio. The scheme diagram is shown in fig. 3a. The interferometer consists of frequency doubler 1 where type I phase matching occurs, polarization-sensitive phase element 2, filter 3 which cuts of SH, and polarizer 4 (or polarizing beam-splitter).

We are interested in phase of the fundamental wave at the exit from the interferometer. Assume the incident wave is linearly polarized and has intensity $I$. Inside the crystal there will be an ordinary wave of intensity
$I \cos^2 \xi$ and extraordinary wave of intensity $I \sin^2 \xi$, where $\xi$ is the angle between the incident polarization and the polarization of the ordinary wave. After the polarizer, which is oriented at 45° with respect to the ordinary wave, there will be interference between a fraction of the extraordinary wave and the corresponding fraction of the ordinary one. Their amplitudes we denote as $a_1^e$ and $a_2^e$, correspondingly. Provided that there is type I phase matching, the generation of SH will decrease the amplitude of only one of the fundamental waves. Let’s assume, for certainty, the ordinary wave is partially converted into the extraordinary wave of SH, and the other fundamental wave (losses neglected) does not vary in intensity. Obviously, the amplitude ratio of the fundamental waves will depend on the input intensity:

$$\frac{a_2^e}{a_1^e} = \left[1 - t h^2(KlI^{1/2} \cos \xi)\right]^{1/2} \frac{\cos \xi}{|\sin \xi|}.$$  \hspace{1cm} (6)

Varying angle $\xi$, we can thus change ratio $a_2^e/a_1^e$, at low intensity.

Expanding (1) with the expression given by (6) we will have the dependence of the resulting wave phase $\Phi$ upon intensity.

The dependence of the nonlinear phase shift of fundamental wave on the incident intensity is demonstrated in fig. 3b, as well as the dependence of output intensity on the incident one.

In the scheme shown in fig. 3a, the nonlinear distortion of the fundamental beam in the output of interferometer (focusing or defocusing) also takes place, which can be controlled by variation of relative phase. The latter is changed by a polarization-sensitive phase element, in this case.

As well as cascaded processes, the described mechanism of ultrafast nonlinearity produces a nonlinear phase shift of a wave without changing refractive index of a medium. These mechanisms of nonlinearity have, essentially, the similar nature. Indeed, NPS occurring at cascading of quadratic processes actually arises from the interference of the incoming fundamental wave and the wave downconverted with a shifted phase from second harmonics. The difference is in the way the interfering waves are produced.

As an analogue to the nonlinear coefficient $n_2$ for cubic nonlinearity, in parallel with second-order cascaded processes, one can introduce the effective refractive index $n_{2\text{eff}} = \Delta \Phi_{\text{NLC}}/\omega I$ for the ”interference” mechanism (two-wave interference with intensity-dependent amplitude ratio). We compare $n_{2\text{eff}}$ for ”interference” phase shift of fundamental wave at $\Delta kl = 0$ to $n_{2\text{eff}}$, related to cascaded second-order processes calculated for optimal mismatch.
$|\Delta k| \approx 3$ [5, 9]. The comparison at $K^2\ell^2 I \leq 10$, that corresponds to $I \leq 22$ GW/cm$^2$ for 1-mm-thick KTP [5], shows that the discussed interference mechanism gives at $\phi_2 - \phi_1 \geq 3\pi/4$ the value of $n^\text{eff}_2$ about $2 \div 3$ times as large as, cascaded second-order processes do, while at $\phi_2 - \phi_1 \approx \pi/2$ these mechanisms produce comparable nonlinear phase shifts.

The proposed technique makes it possible to produce nonlinear phase shift without cascaded second-order processes and cubic nonlinearity. The cascaded processes are eliminated by exact phase matching $\Delta k = 0$ and mutually orthogonal orientation of the crystals. In real materials, $n_2$ for fast cubic nonlinearity is much smaller than $n^\text{eff}_2$ for second-order processes, for instance, in KTP $n_2 \approx 0.1 n^\text{eff}_2$ [9]. Nevertheless, if we allow for inertial cubic nonlinearity ($\tau \approx 1$ ns) [10] for nanosecond pulses $n_2$ exceeds $n^\text{eff}_2$. For experimental observation of the isolated effect in such case one must use picosecond pulses to eliminate the influence of slow nonlinearity.

We have demonstrated a new method for obtaining fast nonlinear shift. The effect has a simple and intuitive interpretation, and is related to interference of two waves with intensity-dependent amplitude ratio caused by second harmonic generation. Sign and value of the nonlinear phase shift is given by the relative phase of the waves introduced by phase element. The effect of "interference" nonlinear phase shift has properties similar to those of the nonlinear phase shift caused by cascaded second-order processes and has several times higher $n^\text{eff}_2$ then the latter. Furthermore, this effect allows producing NPS both of the fundamental and second-harmonic waves.

It worth noting the following features of described technique. As we can see in fig. 3b, the output intensity weakly depends on the input one within a rather wide region. This property seems to be attractive for pulsations damping. The strong dependence of $\Delta \Phi_{NL}$ upon the relative phase near the point $\pi$ (for instance, in fig. 2 $\Delta \Phi_{NL}$ changes from $0.95\pi$ to $-0.95\pi$ as $\phi_2 - \phi_1$ varies within $\pi/25$) can be potentially useful for dispersion interferometry of nonlinear optical media [10, 11].

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Figure Captures

Fig. 1. (a) — The scheme for obtaining nonlinear phase shift in second-harmonic radiation. (b) — The nonlinear phase shift $\Delta \Phi_{NL}$ vs $K^2 l^2 I$; $l_2/l_1 = 2$, $\phi_2 - \phi_1 = 3\pi/4$.

Fig. 2 $\Delta \Phi_{NL}$ (solid line, right axis) and SHG efficiency (dotted line, left axis) vs the relative phase. Here, $l_2/l_1 = 2$, $K^2 l^2 I = 25$
Fig. 3. (a) — The scheme of the device to produce a nonlinear phase shift of the fundamental wave. (b) — The nonlinear phase shift (solid line, right axis) and the output fundamental intensity (dotted line, left axis) vs $K^2t^2I$. Here, $\xi = 0.2$, $\phi_2 - \phi_1 = 3\pi/4$. 
$I_2(\omega) / I$

$\phi_2 - \phi_1$ (units of $\pi$)

$\Delta \phi_{NL}$ (units of $\pi$)
