Rigorous solution to second harmonic generation considering transmission and reflection of light at air–crystal interface

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Considering the transmission and reflection of TE-polarized pump light at the air–crystal interface, the second harmonic generation (SHG) in a lithium niobate (LN) crystal is investigated theoretically and systematically in this work. In previous studies, the theoretical analyses of reflection and transmission of incident wave in the process of nonlinear frequency conversion were not considered in LN crystal on account of the complicated calculations. First, we establish a physical picture describing that a beam of light in TE mode transports in the LN crystal considering transmission and reflection at the crystal surface and generates nonlinear second-order optical polarization in crystal. Then we analytically derive the reflection coefficient and transmission coefficient of pump light by using the dispersion relationships and electromagnetic boundary conditions. We construct the nonlinear coupled wave equations, derive and present the small signal approximation solution and the general large signal solution exactly. Under the transmission model and reflection model, we find that the conversion efficiency of the second-harmonic wave is obviously dependent on transmission coefficient and other general physical quantities such as the length of LN crystal and the amplitude of pump light. Our analytical theory and formulation can act as an accurate tool for the quantitative evaluation of the SHG energy conversion efficiency in an LN crystal under practical situations, and it can practically be used to treat other more complicated and general nonlinear optics problems.

Keywords: nonlinear frequency conversion, transmission, reflection

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1. Introduction

In recent years, tremendous progress in the field of nonlinear optics technology has been made. Considerable effort has been dedicated to generating broadband coherent light sources and ultrafast pulse lasers by using the laser light interacting with nonlinear crystals. Different second-order nonlinear optical effects such as second harmonic generation (SHG), sum frequency generation (SFG), difference frequency generation (DFG), and optical parametric oscillation and amplification allow spectral range to expand to new wavelength band.[1–16] Two schemes called the birefringence phase matching (BPM)[2,17] scheme and quasi-phase-matching (QPM)[3,8] scheme have been proposed to achieve high conversion efficiency. By changing the sign of the second-order nonlinear susceptibility $\chi^{(2)}$ to compensate for the phase mismatching, many periodic,[4,6,18,19] quasi-periodic,[20] chirped,[21] aperiodic[20,22] structures have been designed. More and more crystals are used to interact with lasers to give rise to nonlinear effects.[7,23–25] But, the solution and the discussion for general nonlinear processes considering reflection and transmission of fundamental wave (FW) on the crystal surface are still lacking.

The reflection and transmission of incident wave on crystal surface is a very old and classical problem. The reflection and transmission of incident wave on the crystal surface and the transmission of incident wave through the crystal can be found in the classical textbooks.[26–30] It has been well established that reflection and transmission of light is an indispensable part of linear optics dealing with the light transport across impedance-mismatched air–crystal interface, and in principle it should also be an important subject for nonlinear optics when pump light passes across the surface of nonlinear crystal, thereby triggering off nonlinear frequency conversion. Unfortunately, the extension from the linear optics problem about pump light transport across a nonlinear crystal to nonlinear optics problem of various nonlinear frequency conversion processes (*e.g.*, the second-harmonic generation, SHG) under pump light illuminating nonlinear crystal, with the air–crystal reflection and transmission fully considered, is very rare. Because many nonlinear crystals have a large refractive index, the air–crystal reflection and transmission cannot be ignored, and thus it is highly desirable to develop a systematic, complete, comprehensive theoretical analysis method and process that can quantitatively evaluate spatiotemporal evolution characteristics of various nonlinear frequency conversion processes in the frame of the reflection and transmission model.

In this work, we will take SHG for example and evaluate the SHG efficiency by using the reflection and transmission model. According to Maxwell's equations and the electromagnetic boundary conditions, we will derive a set of analytical equations to describe the whole process when the

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fundamental-wave (FW) pump light polarized in TE mode, whose polarization direction is parallel to the crystal axis, passes across the surface of a z-cut lithium niobite (LN) crystal, is subjected to certain reflection, transmission, and transport in the crystal, and thus inducing the SHG. We want to obtain some explicit analytical mathematical expressions that allow us to directly find out the SHG efficiency depending on various physical parameters in this more complete model considering the reflection and transmission on the crystal surface of LN. This kind of solution method is closer to the practical situation of nonlinear optical experiment, and thus has a better reference significance than the conventional model and method of pure nonlinear optics without considering the air–crystal interface reflection and transmission.

The remainder of this paper is arranged as follows. In Section 2, we introduce the theoretical model of light transport in the LN crystal with considering transmission and reflection of light at air–crystal interface. We exhibit the general physics image of SHG in an LN crystal when transmission and reflection occur. In Section 3, we derive the spatiotemporal evolution of electromagnetic fields of the LN crystal in the TE polarization state, starting from the Maxwell's equations. In Section 4, we analyze the reflection and transmission theory according to the mathematical dispersion relations and the continuous boundary conditions and calculate the reflection coefficient and transmission coefficient of the incident wave. In Section 5, we derive the coupled-wave equations of FW and SHG from wave equation with considering transmission and reflection of light at the air–crystal interface. In Section 6, we further derive and present the exact theoretical analytical solutions with considering the transmission and reflection of incident TE mode at the crystal surface. We then find that the conversion efficiency of SHG depends on some important physical quantities, including the transmission coefficient of incident wave in both small signal approximation model and the general large signal model. In Section 7, We take specific examples to observe intuitively the conversion efficiency of FW to the second-harmonic wave (SHW) by comparing the ideal circumstance without reflection loss with the actual circumstance with reflection loss at the crystal surface. Finally, in Section 8, we summarize the main results of this work and draw some conclusions that the SHGs produced in LN crystal are determined by the reflection and transmission at crystal surface under strict small signal solution and large signal solution situations, and present an overview of the perspective of some new directions worthy of consideration for the study of nonlinear optics.

2. Physical model

Suppose that a monochromatic FW plane-wave beam is incident on a z-cut LN crystal from the air in a coordinate system as shown in Fig. 1(a), where the principal crystalline orientation of LN is collinear with the coordinate axis. More specially, the extraordinary crystalline axis of LN is parallel to the z axis and the ordinary crystalline plane of the crystal is coplanar with the xoy plane in our system. For simplicity, we consider that the ray propagates along the y axis with a TE polarization state which involves the field components (E_z, H_x, H_y). The air has refractive index $n_0 = 1$, relative permittivity $\varepsilon_{\rm r1} = 1$, and relative permeability $\mu_{\rm r1} = 1$. When one focuses on the transport of FW and its conversion into SHW via SHG within the LN crystal, it is convenient to look upon the LN crystal as an infinite crystal, as described in most textbook about nonlinear optics. In the present situation with considering the FW transmission and reflection at the front (first) surface of LN crystal, the crystal can be looked upon as a semi-infinite crystal and the influence from the back (second) surface can be neglected.

The linear permittivity $\varepsilon_{\rm r}$ of the LN crystal can be expressed as

$$\varepsilon_{\rm r} = \begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix} = \begin{pmatrix} n_{\rm o}^2 & 0 & 0 \\ 0 & n_{\rm o}^2 & 0 \\ 0 & 0 & n_{\rm e}^2 \end{pmatrix}, \tag{1}$$

where $\sqrt{\varepsilon_{xx}} = \sqrt{\varepsilon_{yy}} = n_{\rm o}$ and $\sqrt{\varepsilon_{zz}} = n_{\rm e}$ are the refractive indices, respectively, for ordinary light (o-light) and extraordinary light (e-light) in crystal. The incident wave is z-polarized, and corresponds to e-light with $n_{\rm e}$. Moreover, μ_1 is the relative permeability of the LN crystal. More details are explicitly marked in Fig. 1(b).

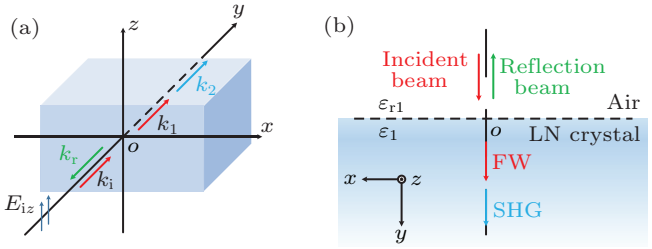


Fig. 1. (a) Schematic diagram of a TE polarization state (polarizes parallel to the z axis) plane wave incident into a z-cut LN crystal in air, described by the wave vectors of incident wave, reflection wave, transmission wave (FW), and SHW. (b) Plan of the propagations of incident wave, reflection wave, transmission wave (FW), and SHW from air to LN crystal.

When the transmission wave at frequency ω_1 propagates through the LN crystal as FW, it will interact with the LN crystal to generate electric polarization $\boldsymbol{P}$, which contains linear and nonlinear terms as follows:

$$\boldsymbol{P} = \varepsilon_0\left(\chi^{(1)} \cdot \boldsymbol{E} + \chi^{(2)} : \boldsymbol{EE} + \chi^{(3)} \vdots \boldsymbol{EEE} + \cdots\right), \tag{2}$$

where $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m is the vacuum permittivity and $\chi^{(1)}$ is known as the linear susceptibility satisfying $\varepsilon_{\rm r} = 1 + \chi^{(1)}$. The quantities $\chi^{(2)}$ and $\chi^{(3)}$ represent the second- and third-order nonlinear susceptibilities, respectively.

The SHG process in the LN crystal originates from the second-order nonlinear polarization $\boldsymbol{P}_{\rm NL}^{(2)} = \varepsilon_0 \chi^{(2)} : \boldsymbol{EE}$. The

FW will excite the nonlinear polarization $\boldsymbol{P}_{\rm NL}^{(2)}$ with frequency $2\omega_{\rm l}$, and $\boldsymbol{P}_{\rm NL}^{(2)} = \boldsymbol{P}_{\rm NL}(2\omega_{\rm l}) = \boldsymbol{P}^{(2)}(2\omega_{\rm l})\mathrm{e}^{-\mathrm{i}\omega_2 t}$ serving as a new radiation source will generate the SHW at frequency $2\omega_{\rm l}$. The nonlinear susceptibility tensor satisfies $d_{il} = \chi^{(2)}/2$ and relies on various FW electric field components and the corresponding nonlinear polarization components, where i and l are contracted Cartesian indices. As a result, the explicit formulation of $\boldsymbol{P}^{(2)}(2\omega_{\rm l})$ for the SHG can be expressed as

$$\boldsymbol{P}^{(2)}(2\omega_{\rm l}) = 2\varepsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \left(E_x^{\omega_{\rm l}}\right)^2 \\ \left(E_y^{\omega_{\rm l}}\right)^2 \\ \left(E_z^{\omega_{\rm l}}\right)^2 \\ 2E_y^{\omega_{\rm l}} E_z^{\omega_{\rm l}} \\ 2E_x^{\omega_{\rm l}} E_z^{\omega_{\rm l}} \\ 2E_x^{\omega_{\rm l}} E_y^{\omega_{\rm l}} \end{pmatrix}. \quad (3)$$

Here, the nonzero nonlinear coefficients are $d_{22} = 2.76$ pm/V, $d_{31} = 5.45$ pm/V, $d_{33} = 27.2$ pm/V in the LN crystal. Note that $E_x^{\omega_{\rm l}}$, $E_y^{\omega_{\rm l}}$, and $E_z^{\omega_{\rm l}}$ are the complex amplitudes of FW field components along the principal coordinate directions x, y, and z, respectively.

3. Electromagnetic solution of TE mode transport

Our analysis is naturally based on the original Maxwell's equations because of the electromagnetic features of light waves. As a consequence, the spatiotemporal evolution[31] of electromagnetic fields with frequency $\omega_{\rm l}$ can be calculated from

$$\nabla \times \boldsymbol{E} = \mathrm{i}\omega_{\rm l}\mu_0\mu_{\rm r}\boldsymbol{H}, \quad \nabla \times \boldsymbol{H} = -\mathrm{i}\omega_{\rm l}\varepsilon_0\varepsilon_{\rm r}\boldsymbol{E}. \quad (4)$$

Here $\mu_0 = 4\pi \times 10^{-7}$ H/m is permeability in vacuum and $\mu_{\rm r}$ is the relative permeability of medium, which is 1 for current situation. Considering TE mode light passing across the surface of LN and transporting along the y axis, equation (4) is reduced to

$$\frac{\partial E_z}{\partial y} = \mathrm{i}\omega_{\rm l}\mu_0\mu_{\rm r}H_x, \quad \frac{\partial E_z}{\partial x} = -\mathrm{i}\omega_{\rm l}\mu_0\mu_{\rm r}H_y,$$
$$\frac{\partial H_x}{\partial y} - \frac{\partial H_y}{\partial x} = \mathrm{i}\omega_{\rm l}\varepsilon_0\varepsilon_{\rm r}E_z. \quad (5)$$

It is found that the electric field for the incident wave, reflection wave, and transmission wave are expressed as

$$E_{\rm iz} = E_{\rm i}\mathrm{e}^{\mathrm{i}k_{\rm iy}y}\mathrm{e}^{-\mathrm{i}\omega_{\rm l}t}, \quad E_{\rm rz} = E_{\rm r}\mathrm{e}^{-\mathrm{i}k_{\rm ry}y}\mathrm{e}^{-\mathrm{i}\omega_{\rm l}t},$$
$$E_{\rm tz} = E_{\rm t}\mathrm{e}^{\mathrm{i}k_{\rm ty}y}\mathrm{e}^{-\mathrm{i}\omega_{\rm l}t}. \quad (6)$$

Here, $E_{\rm i}$, $E_{\rm r}$, and $E_{\rm t}$ denote the amplitude of incident wave, reflection wave, and transmission wave, whose wave vectors are $k_{\rm i} = k_{\rm iy}$, $k_{\rm r} = k_{\rm ry}$, and $k_{\rm t} = k_{\rm ty}$, respectively. Since the incident wave, reflection wave, transmission wave for FW light and SHW light maintain the same polarization direction in the considered model, we can directly calculate and represent the electromagnetic fields in the scalar form.

4. Reflection and transmission of incident wave on LN crystal surface

When the normal incidence occurs, the incident wave, reflection wave, and transmission wave field are expressed in Eq. (6). By substituting Eq. (6) into Maxwell's equation (5), the corresponding magnetic fields can be obtained as follows:

$$H_{\rm ix} = \frac{k_{\rm iy}E_{\rm iz}}{\omega_{\rm l}\mu_0\mu_{\rm r1}}, \quad H_{\rm rx} = -\frac{k_{\rm ry}E_{\rm rz}}{\omega_{\rm l}\mu_0\mu_{\rm r1}}, \quad H_{\rm tx} = \frac{k_{\rm ty}E_{\rm tz}}{\omega_{\rm l}\mu_0\mu_1}. \quad (7)$$

At the same time, we can obtain a series of simple dispersion expressions below:

$$k_{\rm iy}^2 = k_{00}^2\mu_{\rm r1}\varepsilon_{\rm r1}, \quad k_{\rm ry}^2 = k_{00}^2\mu_{\rm r1}\varepsilon_{\rm r1}, \quad k_{\rm ty}^2 = k_{00}^2\mu_1\varepsilon_{zz}^{\omega_{\rm l}}, \quad (8)$$

where $k_{00} = \omega_{\rm l}/c$ indicates the wave vector of incident wave in vacuum, and $\varepsilon_{zz}^{\omega_{\rm l}}$ is the relative permittivity of transmitted FW light in the crystal.

The following continuous boundary conditions are used to describe the matching of tangential components of the electromagnetic field on the interface $y = 0$:

$$E_{\rm iz}|_{y=0} + E_{\rm rz}|_{y=0} = E_{\rm tz}|_{y=0}, \quad H_{\rm ix}|_{y=0} + H_{\rm rx}|_{y=0} = H_{\rm tx}|_{y=0}. \quad (9)$$

Substituting Eqs. (7) and (8) into Eq. (9), and considering that the optical system of homogeneous medium has translational symmetry (described as $k_{\rm iy} = k_{\rm ry}$) in the yoz plane, we can calculate the field reflection coefficient and transmission coefficient when the TE mode with frequency $\omega_{\rm l}$ reaches the LN surface. The result is simply the well-known Fresnel's formula

$$r_{\omega_{\rm l}} = \frac{E_{\rm r}}{E_{\rm i}} = \frac{1 - n_{\rm e}^{\omega_{\rm l}}}{1 + n_{\rm e}^{\omega_{\rm l}}}, \quad (10)$$

$$t_{\omega_{\rm l}} = \frac{E_{\rm t}}{E_{\rm i}} = \frac{2}{1 + n_{\rm e}^{\omega_{\rm l}}}, \quad (11)$$

where $n_{\rm e}^{\omega_{\rm l}} = \sqrt{\mu_1\varepsilon_{zz}^{\omega_{\rm l}}}$ refers to the refractive index of light wave with frequency $\omega_{\rm l}$ in the crystal.

Further, we consider the energy transfer in the presence of reflection and transmission. The energy flux of the wave can be calculated by defining an average pointing vector as follows:

$$\boldsymbol{S} = \frac{1}{2}\mathrm{Re}[\boldsymbol{E}^* \times \boldsymbol{H}]. \quad (12)$$

In this regard, combining Eq. (6) with Eqs. (7) and (12), we have

$$\boldsymbol{S}_{\rm i} = \frac{k_{\rm iy}|E_{\rm i}|^2}{2\omega_{\rm l}\mu_0\mu_{\rm r1}}e_y, \quad \boldsymbol{S}_{\rm r} = -\frac{k_{\rm ry}|E_{\rm r}|^2}{2\omega_{\rm l}\mu_0\mu_{\rm r1}}e_y,$$
$$\boldsymbol{S}_{\rm t} = \frac{k_{\rm ty}|E_{\rm t}|^2}{2\omega_{\rm l}\mu_0\mu_1}e_y. \quad (13)$$

In order to distinguish the pump light from the generated SHW light in the crystal, the field amplitude of the transmitted wave E_t is written as $E_{10}(y)$ and the field amplitude of the generated SHW is denoted as $E_{20}(y)$. The FW energy transmitted to LN crystal is obtained as follows:

$$I_1(y) = |\boldsymbol{S}_t| = \frac{1}{2}\varepsilon_0 c n_1 |E_{10}(y)|^2, \tag{14}$$

where $n_1 = n_e^{\omega_1}$ is the refractive index of FW in the LN crystal.

In the same way, the energy of wave with the other frequency ω_2 is

$$I_2(y) = \frac{1}{2}\varepsilon_0 c n_2 |E_{20}(y)|^2, \tag{15}$$

where $n_2 = n_e^{\omega_2}$ is the refractive index of SHG in the LN crystal. According to Eq. (13), the reflectivity and transmittance of the incident wave are obtained as follows:

$$\begin{aligned} R &= \left|\frac{S_{ry}}{S_{iy}}\right| = |r_{\omega_1}|^2 = \left(\frac{1-n_e^{\omega_1}}{1+n_e^{\omega_1}}\right)^2, \\ T &= \left|\frac{S_{ty}}{S_{iy}}\right| = \frac{k_{ty}}{k_{iy}}\frac{\mu_{r1}}{\mu_1}|t_{\omega_1}|^2 = \frac{4n_e^{\omega_1}}{\left(1+n_e^{\omega_1}\right)^2}. \end{aligned} \tag{16}$$

The expressions of Eq. (16) are the field energy reflection coefficient and transmission coefficient which are related to the refractive index of the FW light polarized in TE mode. The refractive index can be obtained by Sellmeier formula,[32] which shows that it has different values for different wavelengths of the incident wave, namely it has obvious dispersion. As can be seen from Fig. 2, there appear clear changes in the energy reflection coefficient and transmission coefficient with respect to the wavelength in a range of 800 nm–1300 nm, and this is purely attributed to, and thus also reflects the dispersion nature of the LN crystal. As displayed in Fig. 2(a), the reflection coefficient decreases with wavelength increasing monotonically. In contrast, the transmission coefficient increases with wavelength increasing monotonically. As shown in Fig. 2(b), the reflectivity and transmittance of field energy change with the wavelength increasing in a similar trend, which satisfies the law of conservation of energy. From the figure, it is easy to identify the quantity of the reflection coefficient and transmission coefficient, or the reflectivity and transmittance at a fixed incident wavelength of FW pump light. Despite the small variations for different wavelengths, to be more exact, the incident wave energy is reflected by about 14%, while the transmission of wave energy decreases from 100% in the ideal case to about 86% due to the reflection of FW light on the surface of LN crystal, which will yield a noticeable effect on the SHG efficiency. The results are consistent with law of conservation of energy: the more the transmission, the less the reflection is.

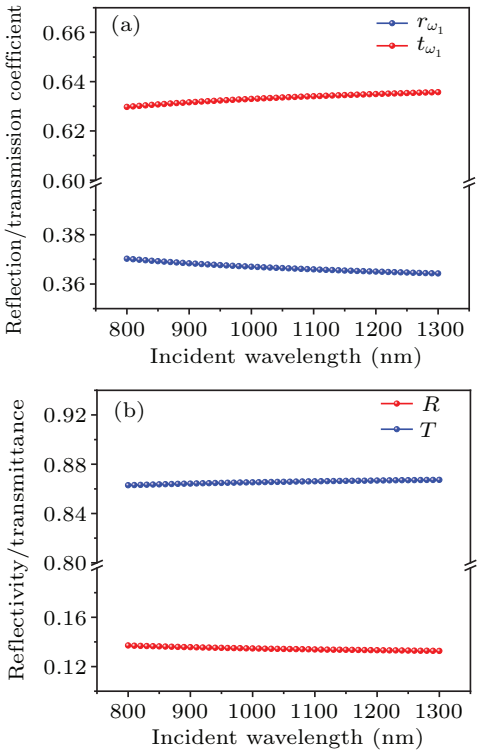


Fig. 2. (a) Calculated reflection coefficient and transmission coefficient of incident wave field amplitude from air to LN crystal surface. (b) Calculated reflectivity and transmittance of incident wave field energy from air to LN crystal surface.

5. Nonlinear coupled-wave theory of SHG in LN crystals

As mentioned above, there are a linear part and a nonlinear part in the polarization P, which leads to the fact that the electric displacement vector D composed of E and P_{NL} also involves two parts. We have

$$D = \varepsilon_0 \varepsilon_r \cdot E + P_{NL}, \tag{17}$$

where ε_r is described by Eq. (1). It is not too difficult to substitute D into Maxwell's equations and reach the following wave equation which describes the evolution behavior of nonlinear wave field in crystal:

$$\nabla^2 E - \mu_0 \varepsilon_0 \varepsilon_r \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}. \tag{18}$$

5.1. Case 1: $\omega_1 + \omega_1 \to \omega_2$

Our derivation starts from the well-established wave equation, which is described by Eq. (18). We first consider the process in which the FW excites the SHW in the crystal. In this case, the wave equation can be rewritten as

$$\nabla^2 E_2 - \mu_0 \varepsilon_0 \varepsilon_r \frac{\partial^2 E_2}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}(2\omega_1)}{\partial t^2}. \tag{19}$$

Here, E_2 denotes the field of SHW, and $P_{NL}(2\omega_1)$ is the nonlinear polarization serving as the secondary radiation source of SHW. Note that we only consider TE polarization mode participating in the nonlinear interaction between FW and SHW.

The forward propagating SHW light has a form of plane wave as

$$E_2 = E_{20}(y)\,\mathrm{e}^{\mathrm{i}k_{2y}y}\,\mathrm{e}^{-\mathrm{i}\omega_2 t}. \tag{20}$$

Here $E_{20}(y)$ is the field amplitude of SHW, varying with the distance slowly; k_{2y} is the propagation wave vector of SHW obtained from $k_{2y} = n_2\omega_2/c$.

Further, we assume the complex amplitude of SHW to be $E_{20}(\omega_2, y)$ in the form of

$$E_{20}(\omega_2, y) = E_{20}(y)\,\mathrm{e}^{\mathrm{i}k_{2y}y}. \tag{21}$$

We use the slowly varying amplitude approximation (SVA)[33] $\partial^2 E_{20}(y)/\partial y^2 \ll k_{2y}\partial E_{20}(y)/\partial y$ to study the amplitude of wave varying very slowly in the range of a single wavelength. Then equation (20) is substituted into the left-hand side of Eq. (19), yielding the resulting equation

$$\nabla^2 E_2 - \mu_0\varepsilon_0\varepsilon_\mathrm{r}\frac{\partial^2 E_2}{\partial t^2} = 2\mathrm{i}k_{2y}\frac{\partial E_{20}(y)}{\partial y}\,\mathrm{e}^{\mathrm{i}k_{2y}y}\,\mathrm{e}^{-\mathrm{i}\omega_2 t}. \tag{22}$$

Again, the wave function for the transmitted FW light is assumed to be

$$E_1 = E_{10}(y)\,\mathrm{e}^{\mathrm{i}k_{1y}y}\,\mathrm{e}^{-\mathrm{i}\omega_1 t}, \tag{23}$$

where $E_{10}(y)$ varying with the distance slowly is the field amplitude of FW, and k_{1y} is the corresponding propagation wave vector in Eq. (23) and calculated from $k_{1y} = k_1 = n_1\omega_1/c$, and $E_{10}(\omega_1, y)$ is the complex amplitude of FW and given by

$$E_{10}(\omega_1, y) = E_{10}(y)\,\mathrm{e}^{\mathrm{i}k_{1y}y}. \tag{24}$$

Notice that the electric field has only the z-component in the process that the normal incident TE propagation mode travels through the LN crystal. From Eq. (3), the induced polarization $\boldsymbol{P}^{(2)}(2\omega_1)$ arising from SHG process can be calculated. There is only z-component in the nonlinear polarization $P_\mathrm{NL}(2\omega_1)$. For convenience, we use the scalar to express $P_\mathrm{NL}(2\omega_1)$ as

$$P_\mathrm{NL}(2\omega_1) = 2\varepsilon_0 d_{33}E_{10}^2(y)\,\mathrm{e}^{\mathrm{i}2k_{1y}y}\,\mathrm{e}^{-\mathrm{i}\omega_2 t}. \tag{25}$$

When this is substituted into the right-hand side of Eq. (19), the result is

$$\mu_0\frac{\partial^2 P^\mathrm{NL}(2\omega_1)}{\partial t^2} = -\frac{2\omega_2^2 d_{33}}{c^2}E_{10}^2(y)\,\mathrm{e}^{\mathrm{i}2k_{1y}y}\,\mathrm{e}^{-\mathrm{i}\omega_2 t}, \tag{26}$$

which leads to the first coupled-wave equation as expressed below:

$$\frac{\partial E_{20}(y)}{\partial y} = \frac{\mathrm{i}d_{33}\omega_2}{n_2 c}E_{10}^2(y)\,\mathrm{e}^{\mathrm{i}\Delta k_y y}, \tag{27}$$

where Δk_y denotes the phase mismatch between FW and SHW, and is defined as

$$\Delta k_y = 2k_{1y} - k_{2y}. \tag{28}$$

Various methods have been developed to overcome the phase mismatch in quadratic nonlinear process.

5.2. Case 2: $\omega_2 - \omega_1 \to \omega_1$

The SHW generated in the nonlinear LN crystal is coupled with the FW to generate a new nonlinear polarization $\boldsymbol{P}_\mathrm{NL}(\omega_1)$, which serves as the new radiation source of FW. The wave equation of the nonlinear wave used in this case is

$$\nabla^2 E_1 - \mu_0\varepsilon_0\varepsilon_\mathrm{r}\frac{\partial^2 E_1}{\partial t^2} = \mu_0\frac{\partial^2 P_\mathrm{NL}(\omega_1)}{\partial t^2}. \tag{29}$$

Here E_1 is the field generated through the coupling between FW and SHW, and has the same form as Eq. (23). The slowly varying amplitude approximation (SVA) $\partial^2 E_{10}(y)/\partial y^2 \ll k_{1y}\partial E_{10}(y)/\partial y$ has been used once again here to solve Eq. (29). The left-hand side of Eq. (29) becomes

$$\nabla^2 E_1 - \mu_0\varepsilon_0\varepsilon_\mathrm{r}\frac{\partial^2 E_1}{\partial t^2} = 2\mathrm{i}k_{1y}\frac{\partial E_{10}(y)}{\partial y}\,\mathrm{e}^{\mathrm{i}k_{1y}y}\,\mathrm{e}^{-\mathrm{i}\omega_1 t}. \tag{30}$$

It is a fairly necessary matter to calculate the nonlinear polarization $\boldsymbol{P}_\mathrm{NL}(\omega_1) = \boldsymbol{P}^{(2)}(\omega_1)\,\mathrm{e}^{-\mathrm{i}\omega_1 t}$.

The complex amplitude $\boldsymbol{P}^{(2)}(\omega_1)$ is given by

$$\boldsymbol{P}^{(2)}(\omega_1) = 4\varepsilon_0 d_\mathrm{il} : \boldsymbol{E}_{20}(\omega_2, y)\,\boldsymbol{E}_{10}(-\omega_1, y).$$

It can be expanded to

$$\boldsymbol{P}^{(2)}(\omega_1) = 4\varepsilon_0\begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \times \begin{pmatrix} 0 \\ 0 \\ E_{20}(\omega_2, y)\,E_{10}(-\omega_1, y) \\ 0 \\ 0 \\ 0 \end{pmatrix}, \tag{31}$$

where the complex amplitude of the SHW field and the FW field come from Eqs. (21) and (24), respectively. And equation (31) brings us $P_\mathrm{NL}(\omega_1)$ in the scalar form as

$$P_\mathrm{NL}(\omega_1) = 4\varepsilon_0 d_{33}E_{20}(y)\,E_{10}^*(y)\,\mathrm{e}^{\mathrm{i}(k_{2y}-k_{1y})y}\,\mathrm{e}^{-\mathrm{i}\omega_1 t}. \tag{32}$$

The right-hand side of Eq. (29) becomes

$$\mu_0\frac{\partial^2 P_\mathrm{NL}(\omega_1)}{\partial t^2} = -\frac{4d_{33}\omega_1^2}{c^2}E_{20}(y)\,E_{10}^*(y)\,\mathrm{e}^{\mathrm{i}(k_{2y}-k_{1y})y}\,\mathrm{e}^{-\mathrm{i}\omega_1 t}. \tag{33}$$

Therefore, the second coupled-wave equation we need has the following simple form:

$$\frac{\partial E_{10}(y)}{\partial y} = \frac{2\mathrm{i}d_{33}\omega_1}{n_1 c}E_{20}(y)\,E_{10}^*(y)\,\mathrm{e}^{-\mathrm{i}\Delta k_y y}. \tag{34}$$

Equations (27) and (34) constitute the coupled-wave equations describing the SHG in LN crystal.

6. Exact solutions of SHG in nonlinear LN crystal under reflection and transmission model

Through the calculation above, we know that when the light in TE polarization mode is normally incident on the surface of the LN crystal, the nonlinear frequency conversion process generated by the transmitted FW light can be completely described by the coupled-wave equations (27) and (34). Now we continue to solve the coupled-wave equations explicitly to establish the quantitative relation of FW and quantitative relation of SHW with the full consideration of the reflection and transmission of FW light at air–crystal interface. There are two kinds of theories dealing with coupled-wave equations, namely small signal approximation theory and general large-signal theory.

6.1. Smallsignal approximation theory of SHG under reflection and transmission model

Under the small-signal approximation theory, we think that the FW pump intensity at frequency ω_1 varies little with transmission distance, so that the intensity (or amplitude) of the pump wave can be treated as an invariant. In other words, the coupling process is considered to be one that does not consume the energy of the pump light. So we have

$$\frac{\partial E_{10}(y)}{\partial y} = 0. \tag{35}$$

By using Eq. (35), we just need to solve Eq. (27) in the coupled wave equations . We can integrate both sides of Eq. (27) from 0 to y, obtaining

$$E_{20}(y) - E_{20}(0) = \frac{\mathrm{i}d_{33}\omega_2}{n_2 c} E_{10}^2(0) \frac{1}{\mathrm{i}\Delta k_y} \left(\mathrm{e}^{\mathrm{i}\Delta k_y y} - 1\right), \tag{36}$$

where y is the length of the crystal.

The initial boundary conditions of FW and SHW are

$$E_{20}(0) = 0, \quad E_{10}(y) = E_{10}(0). \tag{37}$$

The output amplitude of the SHG field can be given by

$$|E_{20}(y)|^2 = \frac{d_{33}^2\omega_2^2 y^2}{n_2^2 c^2} |E_{10}(0)|^4 \operatorname{sinc}^2\left(\frac{\Delta k_y y}{2}\right). \tag{38}$$

Using Eqs. (14) and (15), we can rewrite the connection channels between amplitude and intensity of incident FW, transmitted FW, and generated SHW orderly.

$$I_{00}(0) = \frac{1}{2}\varepsilon_0 n_0 c |E_{00}(0)|^2, \quad I_1(0) = \frac{1}{2}\varepsilon_0 n_1 c |E_{10}(0)|^2,$$
$$I_1(y) = \frac{1}{2}\varepsilon_0 n_1 c |E_{10}(y)|^2, \quad I_2(y) = \frac{1}{2}\varepsilon_0 n_2 c |E_{20}(y)|^2. \tag{39}$$

The definition of transmission coefficient t_{ω_1} from Eq. (11) leads to

$$|E_{10}(0)| = t_{\omega_1} |E_{00}(0)|. \tag{40}$$

Combining Eq. (39) with Eq. (40), the relation between the intensity of incident wave and the transmitted wave can be given as follows:

$$I_1(0) = t_{\omega_1}^2 n_1 I_{00}(0). \tag{41}$$

Therefore, the conversion efficiency of FW to SHW under the reflection and transmission model is determined by

$$\eta = \frac{I_2(y)}{I_{00}(0)} = \frac{8d_{33}^2\omega_1^2 y^2 t_{\omega_1}^4 I_{00}(0)}{\varepsilon_0 n_2 c^3} \operatorname{sinc}^2\left(\frac{\Delta k_y y}{2}\right), \tag{42}$$

which gives us an obvious conclusion that the reflection and transmission of incident wave on LN crystal surface is one of the important factors determining the conversion efficiency of SHG. On the other hand, it is not difficult to see that the conversion efficiency of FW to SHW is dependent on various factors including the transmission distance y, initial intensity $I_{00}(0)$ of the incident wave, and the phase mismatch Δk_y between FW and SHW.

6.2. General large signal theory of SHG under reflection and transmission model

In contrast to the case of the small-signal approximation, there is a consumption of FW energy in general large signal theory. This is the so-called general pump power depletion situation. The energy is converted between the pump FW and the generated SHW. In this case, the pump amplitude cannot be considered as a constant , so

$$\frac{\partial E_{10}(y)}{\partial y} \neq 0. \tag{43}$$

Meanwhile, the initial boundary conditions are

$$E_{10}(0) \neq 0, \quad E_{20}(0) = 0. \tag{44}$$

In this situation, two coupled wave equations in Eqs. (27) and (34) need to be solved simultaneously. We make a discussion on the phase mismatch and the condition for phase matching first. As we have discussed above, the phase mismatch is closely related to the wave vector mismatch, and given by

$$\Delta k_y = \frac{2\pi}{\lambda_2}(n_1 - n_2). \tag{45}$$

Equation (45) indicates that the wavelength (frequency) of incident wave has an effect on the phase mismatch through affecting the refractive index of light in the crystal. The key to achieving the complete phase matching is to meet the equality of the refractive index between FW and SHW in the LN crystal, which unfortunately cannot be realized under the condition of simple normal incidence of TE polarization mode.

However, as a feasible scheme, phase matching can be readily implemented by using the first-order quasi-phase matching (QPM) periodic structure of lithium niobate crystal (PPLN), and the scheme can solve the coupled wave equations by using the general large-signal theory. In this scheme, the

crystal structure is redesigned and the second-order nonlinear polarization coefficient $\chi^{(2)}$ of the crystal is changed so that it can provide the reciprocal lattice vectors (RLVs) exactly equal to Δk_y for compensating for the phase mismatch successfully. The new nonlinear polarization coefficient is called the first-order effective nonlinear polarization coefficient $\chi_{\rm m}$.[34] When the first-order QPM condition is satisfied, $\chi_{\rm m}$ has the value of

$$\chi_{\rm m} = 2\chi^{(2)}/\pi = 4d_{33}/\pi. \tag{46}$$

We define the new phase mismatch quantity $\Delta k_{\rm m}$ as

$$\Delta k_{\rm m} = \Delta k_y - G_{\rm m}, \tag{47}$$

where $\Delta k_{\rm m} = 0$ if the phase matching condition is achieved. In this condition, we can obtain the following nonlinear coupled-wave equations from Eqs. (27) and (34):

$$\frac{\partial E_{10}(y)}{\partial y} = \frac{{\rm i}\omega_1}{n_1 c}\chi_{\rm m} E_{20}(y) E_{10}^{*}(y), \tag{48}$$

$$\frac{\partial E_{20}(y)}{\partial y} = \frac{{\rm i}\omega_1}{n_2 c}\chi_{\rm m} E_{10}^{2}(y). \tag{49}$$

Combing the two equations above, we obtain

$$\frac{\partial |E_{10}(y)|^2}{\partial y} + \frac{n_2}{n_1}\frac{\partial |E_{20}(y)|^2}{\partial y} = 0. \tag{50}$$

We integrate both sides of the equation and make use of the initial boundary conditions from Eq. (44). Finally, we obtain

$$|E_{10}(y)|^2 = |E_{10}(0)|^2 - \frac{n_2}{n_1}|E_{20}(y)|^2. \tag{51}$$

We next take the absolute values of both sides of Eq. (49) and take Eq. (51) into account, then we obtain

$$\frac{\partial |E_{20}(y)|}{\partial y} = \frac{\omega_1}{n_2 c}\chi_{\rm m}\left(|E_{10}(0)|^2 - \frac{n_2}{n_1}|E_{20}(y)|^2\right). \tag{52}$$

Furthermore, we integrate the left side and right side of Eq. (52) and use the integral equation

$$\int \frac{{\rm d}x}{a^2 - x^2} = \frac{1}{a}\tanh^{-1}\left(\frac{x}{a}\right), \tag{53}$$

which gives the output field of the generated SHW as follows:

$$|E_{20}(y)|^2 = \frac{n_1}{n_2}|E_{10}(0)|^2 \tanh^2\left(\frac{\omega_1}{n_2 c}\sqrt{\frac{n_2}{n_1}}\chi_{\rm m}|E_{10}(0)|y\right). \tag{54}$$

Then equations (51) and (54) are then combined and thus yielding

$$|E_{10}(y)|^2 = |E_{10}(0)|^2 \,{\rm sech}^2\left(\frac{\omega_1}{n_2 c}\sqrt{\frac{n_2}{n_1}}\chi_{\rm m}|E_{10}(0)|y\right). \tag{55}$$

Not only the relations listed in Eq. (39) between the intensity and amplitude of wave but also the transmission coefficient t_{ω_1} are utilized to calculate conversion efficiency.

Finally, we find that in the case of the first-order quasi-phase matching, the conversion efficiency of FW to SHW propagation in a nonlinear LN crystal, when considering the air–crystal reflection and transmission, are explicitly expressed as

$$\eta_1 = \frac{I_1(y)}{I_{00}(0)} = n_1 t_{\omega_1}^2 \,{\rm sech}^2\left(\frac{\omega_1}{n_2 c}\sqrt{\frac{n_2}{n_1}}\frac{4d_{33}}{\pi}t_{\omega_1}|E_{00}(0)|y\right),$$

$$\eta_2 = \frac{I_2(y)}{I_{00}(0)} = n_1 t_{\omega_1}^2 \tanh^2\left(\frac{\omega_1}{n_2 c}\sqrt{\frac{n_2}{n_1}}\frac{4d_{33}}{\pi}t_{\omega_1}|E_{00}(0)|y\right). \tag{56}$$

It is quite clear that the form of the rigorous solution contains the transmission coefficient of incident FW light, and this verifies that the reflection and transmission of incident wave cannot be neglected.

7. Results and discussion

7.1. Small signal solution of SHG under reflection and transmission model

We discuss and analyze the small signal solution of Eq. (42) of SHG under the reflection and transmission model by considering a practical example. For comparison, an LN crystal and a PPLN with first-order quasi-phase matching structure are explored. In our example, the wavelength and amplitude of the incident pump FW light beam are set to 800 nm and $E_{00} = 4\times10^6$ V/m. When the light beam is incident from the air into LN crystal, the corresponding refractive index in crystal is 2.176 for the FW and 2.332 for the SHW. In crystal, the phase mismatch between FW and SHW is equal to 2.4488 $\mu{\rm m}^{-1}$ where the wavelength is set to 800 nm so that shorter distance is selected to clearly observe the oscillatory characteristics of the curves. We will explore the intensity of light at a defined position in a crystal. The propagation length of LN is set to 0.1 mm for LN and 0.8 mm for PPLN. We investigate the evolution of the SHW conversion efficiency η with the propagation distance y, and comparisons are made between the models without and with considering the air–crystal reflection and transmission of pump FW light denoted with lines in different colors, as illustrated in Figs. 3(a) and 3(b), respectively.

As can be seen from Fig. 3, according to the small signal approximation theory, the energy conversion efficiency of SHW in LN crystal changes with the propagation distance, showing the characteristic of periodic oscillation due to phase mismatching, while the energy conversion efficiency of SHW in PPLN crystal presents a monotonically increasing trend with the propagation distance as the phase mismatching vanishes. Yet, the differences in the quantity of conversion efficiency between the two models under the ideal condition and the actual condition are distinctly observable in both LN and PPLN.

Firstly, in the LN crystal, the curve profiles resemble the squared term of the sinc function in Eq. (42). The amplitude of the SHW efficiency depends on the wavelength if the incident light intensity and crystal length are both fixed in the analysis model, while the oscillation period relies on the phase mismatch determined by the wavelength essentially and the propagation length as well. Moreover, we can find an optimal length for the LN crystal interacting with optical wave to evaluate the highest SHG conversion efficiency. The feature can be disclosed that the influence from reflection will not bring about the period change of oscillation. Secondly, it is apparent that the conversion efficiency of the SHW decreases from 1.76×10^{-7} to 1.31×10^{-7} for LN crystal (both obviously are very small, Fig. 3(a)) but from 6.86% to 5.11% for QPM PPLN crystal (both are modestly large, Fig. 3(b)) when the laser beam transports a distance of 0.8 mm within the crystal due to the energy consumed for the reflection on the crystal surface in LN crystal at the wavelength of 800 nm. It implies that reflection and transmission cannot be neglected in the discussion on nonlinear frequency conversion of pump light wave. This example gives an intuitive way to confirm that the air–crystal transmission coefficient plays an important role in manipulating SHG as an indispensable optical parameter.

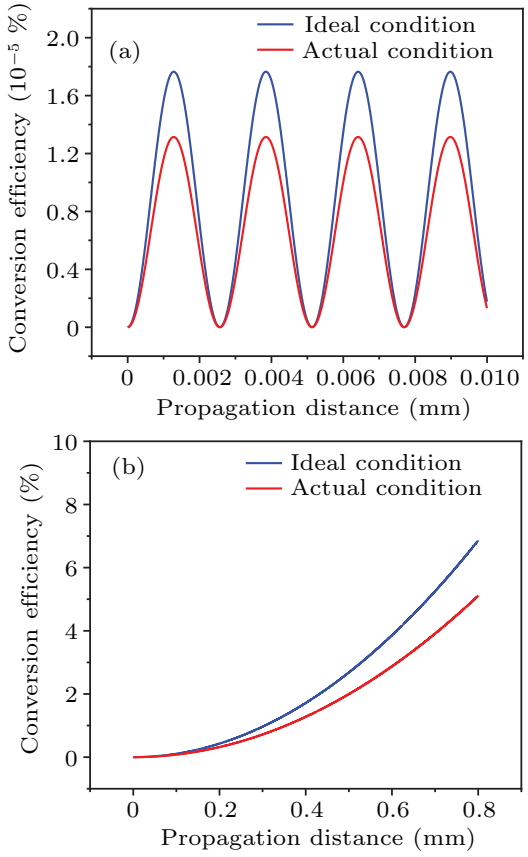


Fig. 3. Conversion efficiency of FW to SHW under the small signal approximation theory on (a) LN crystal surface where reflection and transmission are ignored in ideal condition and considered in actual condition; (b) the first-order quasi-phase matching PPLN crystal surface where reflection and transmission are ignored in ideal condition and considered in actual condition.

7.2. General large signal solution of SHG under reflection and transmission model

Our discussion on large signal solution starts from the analytical expressions of Eq. (56). In order to highlight the effect of reflection and transmission of incident wave more intuitively, we compare the models without and with considering reflection and transmission of wave on the PPLN crystal surface as seen in Figs. 4(a) and 4(b), respectively. It should be noted that the crystal under consideration has a first-order QPM structure. Here the incident wave has a wavelength of 800 nm, generating SHW at 400 nm in the PPLN crystal. We select $y = 10$ mm to discuss the changes of the FW intensity. The amplitude of incident wave is $E_{00} = 4 \times 10^6$ V/m.

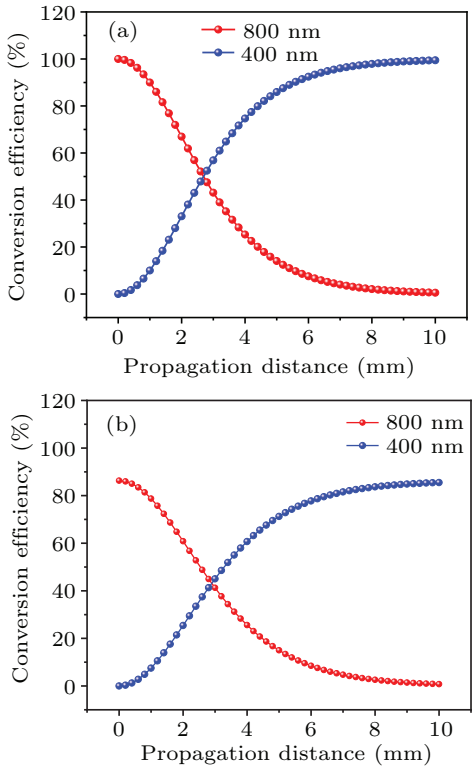


Fig. 4. Conversion efficiency between FW and SHW in general large signal theory on the first-order quasi-phase matching PPLN crystal surface without (a) and with (b) considering reflection and transmission.

Unlike the case of the small signal solution, the FW can be converted into the SHW significantly in the current large signal solution when the phase matching condition is satisfied. For the case that the reflection effect on the crystal surface is negligible, the energy of FW tends to transfer to the generated SHW fully if the FW propagates about 10 mm. However, for the case that reflection effect is considered, about 86.3% energy can be converted if the FW propagates the same distance. The point where the FW energy equals the SHW energy has a right shift because of the role of optical wave reflection. The comparison between the two figures indicates that under the condition of first-order quasi-phase matching, the FW needs to travel a longer distance in the nonlinear crystal material to

realize entirely nonlinear frequency conversion due to the reflection and transmission of incident wave on the PPLN surface. The reflection phenomenon brings about spatial delayed response in the SHW coupling process.

8. Conclusions and perspective

In this work, we present a series of rigorous solution of coupled wave equations for SHG, fully considering the reflection and transmission for FW pump light passing through the air–LN crystal interface. Both the small-signal approximation solution and the general large-signal solution are discussed. The results indicate that the conversion efficiency of SHG depends on transmission coefficient of incident wave visibly and other general physical quantities. It should be noticed that in many previous researches of nonlinear optics, such a reflection and transmission effect was neglected, thereby greatly simplifying the solution of nonlinear optics. Yet, our present theoretical solution indicates that considering the reflection and transmission at the air–crystal interface does not add any extra burden to nonlinear optics problems.

Our theoretical solution involves several major steps. Firstly, we consider the electromagnetic boundary conditions and translational symmetry of homogeneous medium, and obtain both the reflection coefficient and transmission coefficient of field amplitude, reflectivity and transmittance of field energy. Secondly, under the reflection and transmission model, our derivation starts from the electromagnetic field wave equations rooting in Maxwell's equations and we obtain the nonlinear coupled-wave equations. The phase mismatching is also calculated and discussed. Thirdly, under the reflection and transmission model, we derive the small-signal approximate theory and the general large-signal theory from the coupled-wave equations. We obtain the small-signal approximation solution and general large-signal solution about transmission coefficient, where the conversion efficiency of the SHW is regulated by the transmission coefficient. It directly reveals that the relation between the nonlinear frequency conversion and the reflection and transmission of light wave.

We use the new theory to treat several practical examples, which show that the reduction of incident wave energy, caused by the air–crystal reflection effect, leads the SHW conversion efficiency to decrease. We compare the situations where the reflection and transmission are considered and neglected respectively, and find that the light loss caused by the first air–crystal surface reflection is significant and observable. Note that our model can be directly extended and applied to treating the second crystal–air interface when light goes across a crystal. These practical examples intuitively highlight the negative effects of reflection and transmission on nonlinear process. Our work clearly show that the transmission coefficient of incident wave is an important index to evaluate the conversion between FW and SHW in the LN crystal and cannot be ignored. Our new physical model and the analytical theory are clear, simple, and easy to use. At the same time, our work also proposes a new direction worthy of consideration for the study of nonlinear optics. In future studies, the current theory developed for the simple TE mode (the scalar-wave nonlinear optics for LN crystal) can be extended to treating the TM mode nonlinear optics, which is more complex and has more polarization components, for exploring and achieving a more complete theoretical system model. Moreover, in addition to LN crystals, there are other popular and widely used crystals, such as barium borate (BBO), lithium tantalite ($LiTaO_3$), *etc*., which are also worth studying.

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