

# Cascaded wavelength conversion as favorable application of nonlinear optical polymers

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**Abstract:** Nonlinear optical (NLO) polymers have been considered promising materials for wavelength conversion at a low pump power. However, they have not been readily adopted to practical applications due to their high absorption coefficients, especially at a shorter interacting wavelength. Our theoretical analysis proves that the influence of absorption coefficients can be mitigated significantly in cascaded wavelength conversion (CWC) processes. According to our example study, maximum conversion efficiencies for CWC can compare even with those for second-harmonic generation in many NLO polymers. Thus CWC can become a pertinent application of NLO polymers. However, to obtain such efficient CWC, several realistic problems should be resolved in practical devices.

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## 1. Introduction

Optical polymers present great advantages for the development of various optical communication devices by grace of the wide spectrum of their material properties [1, 2]. They also can acquire very high second-order optical nonlinearities by attaching prominent nonlinear optical (NLO) chromophores and then aligning molecular dipoles using an electric poling process. Such NLO polymers typically exhibit ultrafast optical response times. Thus, they have been regarded as excellent materials for efficient and ultrafast electro-optic (EO) or NLO waveguide

devices. For instance, EO devices with very low half-wave voltages were successfully demonstrated with NLO polymers [3, 4].

However, those polymers have hardly been accepted into practical NLO wavelength conversion (WC) devices. This is mainly due to their high optical absorption coefficients at a shorter interacting wavelength region, e.g., at a second-harmonic (SH) wavelength [5, 6], and also the material deterioration effects induced by the high absorption [7, 8]. The high absorption coefficient at the shorter wavelength region originates from the tails of the absorption peaks of NLO chromophores [5]. Thus, it has not been achieved ordinarily to lower the absorption without sacrificing the high nonlinearity owing to the intrinsic trade-off relationship between them. Instead, peculiar phase-matching (PM) schemes were proposed to alleviate the absorption effect [5, 9]. However, there still remain various obstacles to realize practical efficient WC devices with these schemes.

For several years, our group has developed a number of NLO polymers applicable to practical EO or NLO devices. They possess favorable nonlinear optical properties along with superior thin-film quality and thermal stability [10–15]. Moreover, some of them exhibit relatively low absorption coefficients at both fundamental and SH regions ( $\sim 1.55 \mu\text{m}$  and  $\sim 0.78 \mu\text{m}$ , respectively) of an optical communication wavelength [11, 15]. Based on these NLO polymers, we have demonstrated remarkable conversion efficiencies in quasi-phase-matched (QPM) SH generation (SHG) experiments [15, 16]. In the meantime, we have reported our comprehensive analyses on the influence of absorption and nonlinearity in NLO wavelength conversion, and have disseminated our approach to the development of practical NLO polymers with a superb combination of nonlinear and absorption coefficients [6, 17].

In recent years, we have also paid our attention to cascaded WC (CWC) [17–20] because it may become an important application area of NLO polymers in the field of optical fiber communications. In this application, an SH wave at  $\omega_{sh}$  is generated from the frequency doubling of an intense pump wave at  $\omega_p$ , and is successively coupled with a signal wave at  $\omega_s$  to generate a converted (or duplicate) signal wave at  $\omega_c (= 2\omega_p - \omega_s)$  via difference-frequency generation (DFG). This scheme can be utilized as a feasible WC technique for wavelength-division multiplexed optical networks in that the frequencies of  $\omega_p$ ,  $\omega_s$ , and  $\omega_c$  can be closely placed in the 1.55- $\mu\text{m}$  band. Furthermore, the high absorption coefficients of typical NLO polymers at  $\omega_{sh}$  may not affect significantly the essential conversion efficiency from the signal wave to the converted wave, for the SH wave plays a role of a stepping-stone between the two successive second-order NLO processes.

In this paper, we analyze theoretically the effects of the absorption coefficients and optical nonlinearity on the CWC process under the undepleted-pump regime. We utilize the analytical results directly to evaluate several NLO polymers by estimating and comparing their maximum achievable CWC efficiencies, and demonstrate that the CWC process can be applied as a potential application of NLO polymers. We also discuss several realistic problems encountered in our development of practical NLO WC devices.

## 2. Theoretical Analysis

### 2.1. Loss Effect on Conversion Efficiency

Our interest is focused only on the CWC process under the condition of phase-matched SHG ( $\omega_p + \omega_p \rightarrow \omega_{sh}$ ). In this case, the DFG process ( $\omega_{sh} - \omega_s \rightarrow \omega_c$ ) also satisfies its PM condition practically if  $\omega_s$  and  $\omega_c$  are placed very near to  $\omega_p$ . For example, a 3-dB acceptance bandwidth for CWC of  $\sim 60 \text{ nm}$  can be typically obtained with a 5-cm long periodically poled LiNbO<sub>3</sub> crystal under a phase-matched SHG condition [18, 20]. Therefore, we can develop our analysis without regard to phase-mismatch factors for those NLO interactions involved dominantly. Then, including the terms related to the absorption loss at each wavelength, the governing

coupled-amplitude equations can be written in the form

$$\frac{dA_l}{dz} = -\frac{\alpha_l}{2}A_l + \frac{i\omega_l}{n_l c} \sum_{(m,n)} d_{\text{eff}} A_m A_n. \quad (1)$$

Here the subscripts  $l$ ,  $m$ , and  $n$  indicate the subscripts  $p$ ,  $sh$ ,  $s$ , or  $c$ .  $A_l \equiv A(\omega_l)$  represents the complex amplitude of the electric field at the angular frequency component  $\omega_l$ , and satisfies the relation of  $A(-\omega_l) = A^*(\omega_l)$ .  $\alpha_l$  is the absorption coefficient at  $\omega_l$  and  $d_{\text{eff}}$  is the effective second-order NLO coefficient. The summation is to be performed over all the ordered pairs of  $(m, n)$  that satisfy  $\omega_l = \omega_m + \omega_n$ . We here disregard the effect of third-order optical nonlinearity for simplicity.

If we adopt the undepleted-pump assumption given by

$$|A_p| \gg |A_s|, |A_{sh}| \gg |A_c|, \quad (2)$$

the intensities of the pump and signal waves can be simply represented in the form of  $I_l(z) = I_l(0)\exp(-\alpha_l z)$ , where  $l = p$  or  $s$ . We also obtain the intensities of the SH and converted waves expressed as

$$I_{sh}(z) = \frac{8\pi^2 d_{\text{eff}}^2}{c\epsilon_0 n_p^2 n_{sh} \lambda_p^2} I_p^2(0) z^2 \mathcal{L}_{\text{SHG}}(z) \quad (3)$$

and

$$I_c(z) = \frac{16\pi^4 d_{\text{eff}}^4}{c^2 \epsilon_0^2 n_p^2 n_{sh}^2 n_s n_c \lambda_p^2 \lambda_c^2} I_p^2(0) I_s(0) z^4 \mathcal{L}_{\text{CWC}}(z) \quad (4)$$

under the situation of type-I SHG PM and nondegenerate DFG.  $\mathcal{L}_{\text{SHG}}(z)$  and  $\mathcal{L}_{\text{CWC}}(z)$  are the loss factors for phase-matched SHG and CWC processes, respectively, that are given by

$$\mathcal{L}_{\text{SHG}}(z) = \exp[-(\alpha_{sh} + 2\alpha_p)z/2] \left\{ \frac{\sinh[(\alpha_{sh} - 2\alpha_p)z/4]}{(\alpha_{sh} - 2\alpha_p)z/4} \right\}^2 \quad (5)$$

and

$$\begin{aligned} \mathcal{L}_{\text{CWC}}(z) &= \frac{\exp(-\alpha_c z)}{[(\alpha_{sh} - 2\alpha_p)z/4]^2} [g((\alpha_{sh} + \alpha_s - \alpha_c)z) - g((2\alpha_p + \alpha_s - \alpha_c)z)]^2 \\ &\cong \frac{\exp(-\alpha_p z)}{[(\alpha_{sh} - 2\alpha_p)z/4]^2} [g(\alpha_{sh} z) - g(2\alpha_p z)]^2, \end{aligned} \quad (6)$$

where  $g(x)$  is defined by

$$g(x) \equiv \exp(-x/4) \frac{\sinh(x/4)}{x/4}. \quad (7)$$

In the second line of Eq. (6), we suppose  $\alpha_p \cong \alpha_s \cong \alpha_c$  because our interest converges on the CWC process in the 1.55- $\mu\text{m}$  optical communication band. We note here that the result in Eq. (6) is slightly different with that in Ref. [17], which was miswritten by our mistake. Although the above results are based directly on type-I SHG PM, they can be easily adopted to the CWC process involving type-II SHG PM as in Ref. [6].

Figure 1 exhibits the dependence of  $\mathcal{L}_{\text{SHG}}(z)$  and  $\mathcal{L}_{\text{CWC}}(z)$  on the absorption coefficients at the pump and SH waves, in which we adopt the parameters  $\alpha'_p z$  and  $\alpha'_{sh} z$  with the unit of dB, instead of  $\alpha_p z$  and  $\alpha_{sh} z$ . Throughout this paper, we represent the absorption coefficients in both forms of  $\alpha_l$  and  $\alpha'_l$  ( $l = p$  or  $sh$ ). However, the use of  $\alpha_l$  is restricted to analytic expressions only, since the unit of dB/cm is typically employed for absorption coefficients in practical

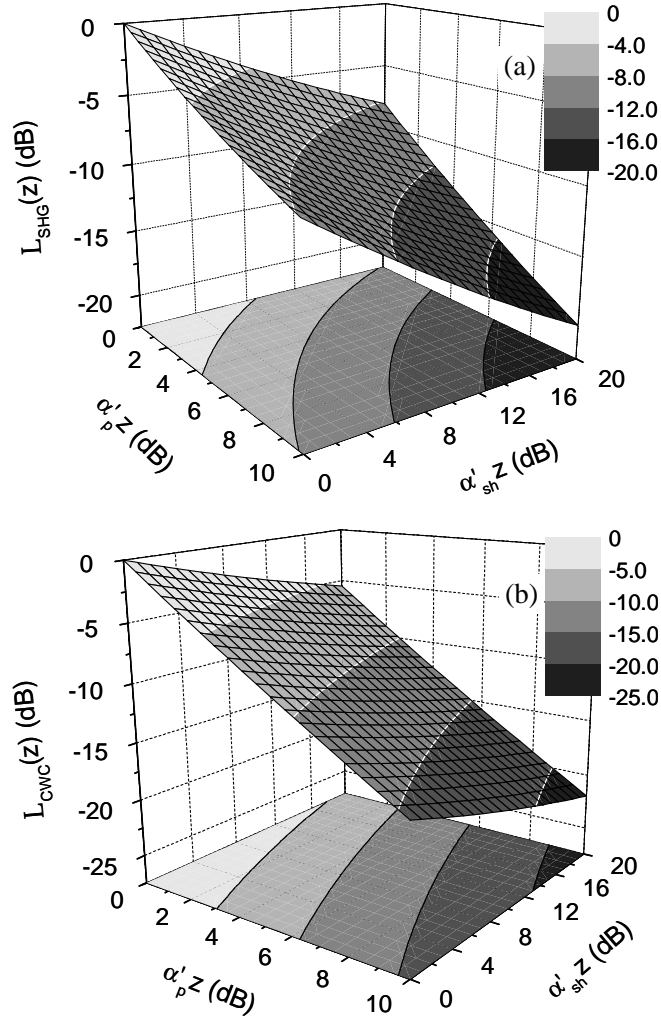


Fig. 1. Loss factors for phase-matched (a) SHG and (b) CWC processes as functions of the absorption coefficients at the pump and SH waves.

situations. Both parameters are related by the expression  $\alpha_l = (\alpha'_l \ln 10)/10$  and 1 dB/cm is nearly equivalent to  $0.23 \text{ cm}^{-1}$ .

It must be noted that Fig. 1(a) is identical to the result for  $\mathcal{D}(z)$  in Ref. [6], which was obtained under the phase-matched SHG condition and was termed the diminution factor. This identical result is a direct consequence of the assumption  $|A_{sh}| \gg |A_c|$  given in Eq. (2). Therefore, we will not include in this paper all the relevant results for the SHG process that are the same as those in Ref. [6]. From Fig. 1(b), we can find that  $\mathcal{L}_{\text{CWC}}(z)$  shows somewhat insensitive dependence on  $\alpha'_{sh}z$ , as we expected. On top of that, the loss effect in the CWC process is rather insignificant than in the SHG process when  $\alpha'_{sh} > 2\alpha'_p$ .

Further qualitative comparison must be made with the conversion efficiencies for the SHG and CWC processes that are defined as  $\eta_{\text{SHG}}(z) \equiv I_{sh}(z)/I_p(0)$  and  $\eta_{\text{CWC}}(z) \equiv I_c(z)/I_s(0)$ , respectively. A simple substitution leads us to obtain the relationship between  $\eta_{\text{SHG}}(z)$  and

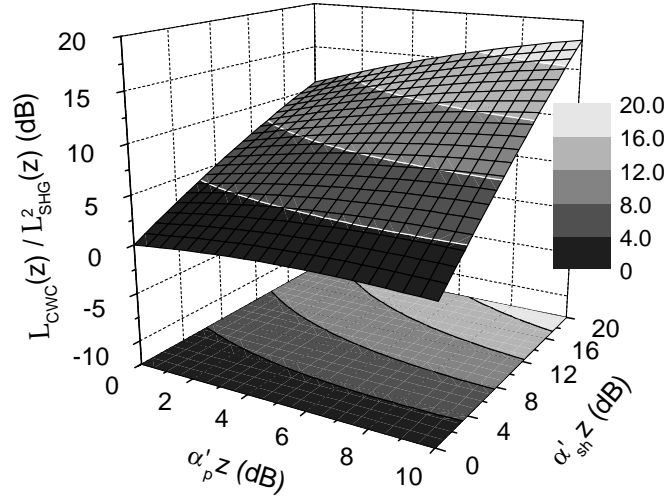


Fig. 2. The benefit factor of the CWC process, relative to the SHG process, as a function of absorption coefficients.

$\eta_{\text{CWC}}(z)$  as

$$\eta_{\text{CWC}}(z) \cong \frac{1}{4} \eta_{\text{SHG}}^2(z) \frac{\mathcal{L}_{\text{CWC}}(z)}{\mathcal{L}_{\text{SHG}}^2(z)}. \quad (8)$$

If all the absorption coefficients can be ignorable, both  $\mathcal{L}_{\text{SHG}}$  and  $\mathcal{L}_{\text{CWC}}$  are given by 1. Then, we can estimate the value of  $\eta_{\text{CWC}}$  from a known value of  $\eta_{\text{SHG}}$  using the simple relationship of  $\eta_{\text{CWC}} \cong \eta_{\text{SHG}}^2/4$ . However, when the absorption coefficients have unignorable values, we must consider the additional factor given by  $\mathcal{L}_{\text{CWC}}(z)/\mathcal{L}_{\text{SHG}}^2(z)$ . The dependence of this factor on the absorption coefficients is plotted in Fig. 2. The factor has the value of 1 in the lossless situation and yields values higher than 1 in all other practical situations. For this reason,  $\mathcal{L}_{\text{CWC}}(z)/\mathcal{L}_{\text{SHG}}^2(z)$  can be identified as the benefit factor of the CWC process, in comparison to the SHG process, in the presence of absorption losses. In particular, we can obtain a high benefit factor at a large value of  $\alpha'_{sh}z$ , while the values are near 1 when  $\alpha'_{sh}z < 1$ . This result evidently reflects the advantageous aspect of the CWC process in an NLO medium with a large  $\alpha'_{sh}$  value.

## 2.2. Maximum CWC Efficiency

Provided the loss effect is not ignorable, the intensities of the SH and converted waves do not increase monotonously as the NLO interaction length increases. Therefore, we take interest in the maximum available value of the conversion efficiency for the development of a practical CWC device. As expressed in Eqs. (3) and (4), we must consider the explicit effect of the NLO interaction length as well as the loss factor to obtain the maximum CWC efficiency.

Although the NLO and absorption coefficients cause conflicting effects on the CWC efficiency, the optimum interaction length for the maximum CWC efficiency is determined solely by the absorption coefficients as in the SHG process [6]. Based on Eq. (4), we can obtain the optimum CWC interaction length  $z_{\text{CWC}}^{\text{max}}$  from the condition of  $d[z^4 \mathcal{L}_{\text{CWC}}(z)]/dz = 0$ . However,  $z_{\text{CWC}}^{\text{max}}$  can not be simply expressed by a definite form. Instead, it is possible to obtain its values by solving numerically the relation of

$$\left(1 + \frac{\alpha_p}{\alpha_{sh}}\right) \exp(-\alpha_{sh} z_{\text{CWC}}^{\text{max}}/2) - \frac{\alpha_p}{\alpha_{sh}} = \frac{3}{2} \exp(-\alpha_p z_{\text{CWC}}^{\text{max}}) - \frac{1}{2}. \quad (9)$$

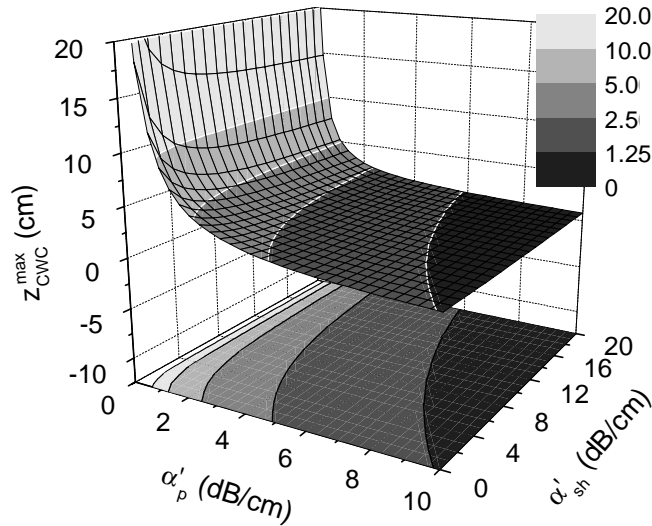


Fig. 3. The optimum CWC interaction length calculated numerically as a function of the absorption coefficients.

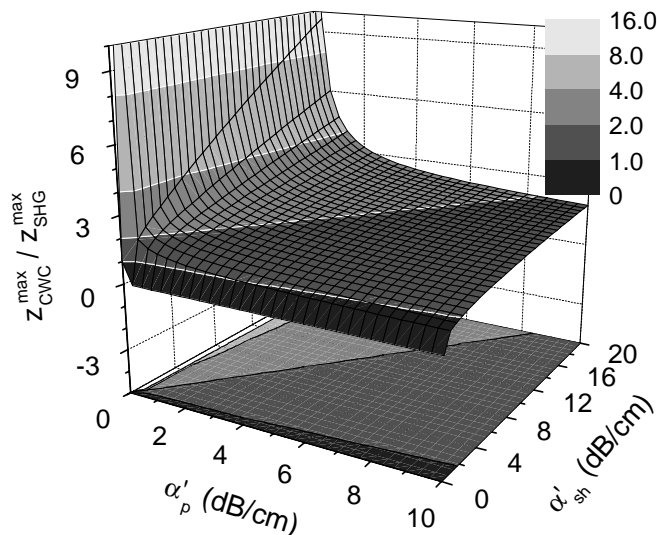


Fig. 4. The ratio between the optimum CWC and SHG interaction lengths as a function of the absorption coefficients.

Though, it must be considered that Eq. (9) can not yield a definite solution when  $\alpha_{sh} = 2\alpha_p$ , and Eq. (9) is also satisfied by the trivial solution of  $z_{\text{CWC}}^{\text{max}} = 0$ , where the CWC efficiency actually has the minimum value. Figure 3 shows the calculated values of  $z_{\text{CWC}}^{\text{max}}$ . We can notice from this result that a little decrease of  $\alpha'_p$  gives rise to a considerable increase in  $z_{\text{CWC}}^{\text{max}}$  in the region of  $\alpha'_p \leq 2$  dB/cm, while  $\alpha'_{sh}$  yields much insensitive influence on  $z_{\text{CWC}}^{\text{max}}$ .

In Fig. 4, we exhibit the result of comparison between the optimum CWC and SHG interaction lengths. The numerical comparison leads us to find that  $z_{\text{CWC}}^{\text{max}}$  is greater than the optimum SHG interaction length  $z_{\text{SHG}}^{\text{max}}$  if  $\alpha'_{sh} > \alpha'_p / 5.65$ . Considering that the NLO interaction length

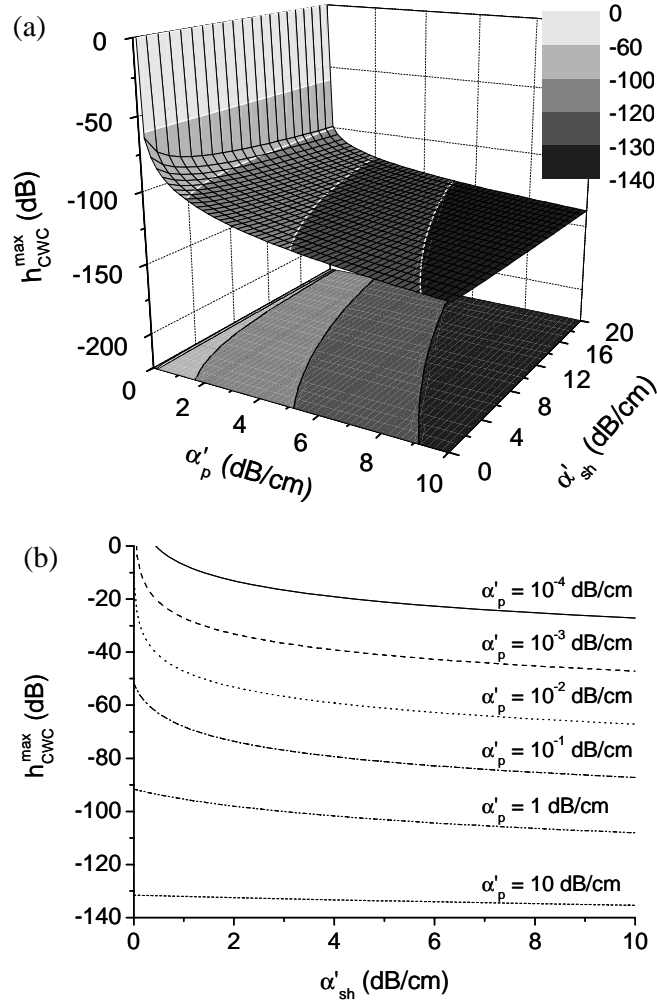


Fig. 5. The coefficient  $h_{\text{CWC}}^{\text{max}}$  calculated for a typical NLO polymer at  $\lambda_p = 1.55 \mu\text{m}$  and  $\lambda_s = 1.56 \mu\text{m}$  when the values of  $d_{\text{eff}}$  and  $I_p(0)$  are given in the units of pm/V and  $\text{kW}/\text{cm}^2$ , respectively: (a) a three-dimensional contour map and (b) a two-dimensional plot for several specific  $\alpha'_p$  values.

is involved in the CWC efficiency with quartic dependence, rather than quadratic dependence as in the SHG conversion efficiency, the increased optimum interaction lengths in that region result in significantly enhanced CWC efficiencies compared with available SHG conversion efficiencies.

With the calculated values of  $z_{\text{CWC}}^{\text{max}}$ , maximum CWC efficiencies can be obtained from the definition of  $\eta_{\text{CWC}}^{\text{max}} \equiv I_c(z_{\text{CWC}}^{\text{max}})/I_s(0)$ . Then we can express the result as

$$\eta_{\text{CWC}}^{\text{max}} = h_{\text{CWC}}^{\text{max}}(\alpha_p, \alpha_{sh}) d_{\text{eff}}^4 I_p^2(0), \quad (10)$$

or equivalently as

$$\eta_{\text{CWC}}^{\text{max}} (\text{in dB}) = h_{\text{CWC}}^{\text{max}} (\text{in dB}) + 40 \log d_{\text{eff}} + 20 \log I_p(0) \quad (10')$$



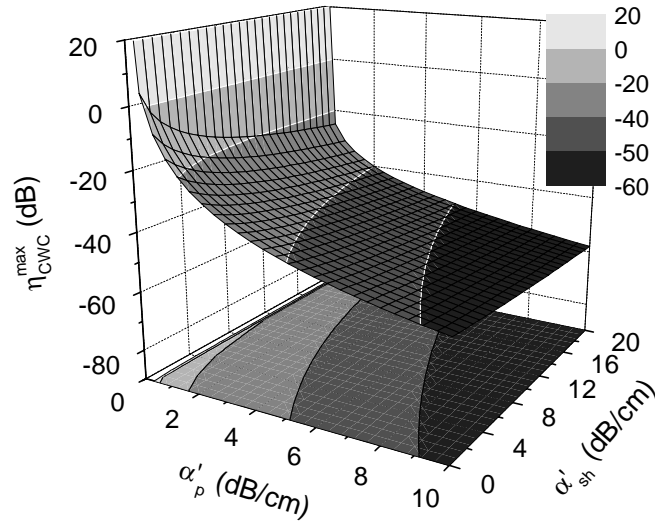


Fig. 6. The maximum CWC efficiency calculated under the condition of  $d_{\text{eff}} = 10 \text{ pm/V}$  and  $I_p(0) = 100 \text{ kW/cm}^2$ .

in the unit of dB, where we represent separately the dependence on the NLO coefficient and the pump intensity. The coefficient  $h_{\text{CWC}}^{\text{max}}$  includes the effects of all the linear optical parameters of a given material. However, it can be simply regarded as just a function of absorption coefficients because other parameters vary only within limited ranges in various NLO polymers for the CWC process of a wavelength in the  $1.55\text{-}\mu\text{m}$  band.

Figure 5 shows the values of  $h_{\text{CWC}}^{\text{max}}$  calculated with the refractive index data of a PEI-DASS polymer [a polyetherimide-based side-chain polymer with a kind of 4-(dialkylamino)-4'-(styrylsulfonyl)-stilbene chromophore], which was developed in our laboratory. The calculation is carried out with  $\lambda_p = 1.55 \text{ }\mu\text{m}$  and  $\lambda_s = 1.56 \text{ }\mu\text{m}$ , provided that the values of  $d_{\text{eff}}$  and  $I_p(0)$  are given in the units of  $\text{pm/V}$  and  $\text{kW/cm}^2$ , respectively. The calculated result reconfirms that  $\alpha'_p$  makes a decisive effect on the CWC efficiency, while the effect of  $\alpha'_{sh}$  is rather insignificant. Upon closer examination with Fig. 5(b), it is found that the dependence on  $\alpha'_p$  is expressed by  $h_{\text{CWC}}^{\text{max}}$  (in dB)  $\propto -40 \log \alpha'_p$  at  $\alpha'_{sh} = 0 \text{ dB}$ , and shows the tendency to approach the relation of  $h_{\text{CWC}}^{\text{max}}$  (in dB)  $\propto -20 \log \alpha'_p$  as the ratio  $\alpha'_{sh}/\alpha'_p$  increases. In the meantime, the effect of  $\alpha'_{sh}$  is represented quite well by rational equations, and also can be described by linear equations in wide range of  $\alpha'_{sh}$  when  $\alpha'_p \gg 1 \text{ dB/cm}$ .

Figure 6 presents the values of  $\eta_{\text{CWC}}^{\text{max}}$  calculated under the condition of  $d_{\text{eff}} = 10 \text{ pm/V}$  and  $I_p(0) = 100 \text{ kW/cm}^2$ , as a typical example. We note here that, contrary to the case of the SHG process, it is possible for the CWC process to obtain a conversion efficiency higher than 0 dB, which corresponds to the conversion efficiency of 100%, in the region of  $\alpha'_p < 0.5 \text{ dB/cm}$ . Moreover, the region of such high conversion efficiencies can be rather extended if the values of  $d_{\text{eff}}$  and  $I_p(0)$  are increased. This is not a strange fact in the CWC process because the pump wave is different from the signal wave and, thus,  $I_s(z)$  can also have a much higher value than  $I_s(0)$  as  $I_c(z)$  increases.

As mentioned above, the CWC efficiency can be estimated from the relation of  $\eta_{\text{CWC}} \cong \eta_{\text{SHG}}^2/4$  when the absorption coefficients can be ignorable. Thus the ratio of  $4\eta_{\text{CWC}}^{\text{max}}/\eta_{\text{SHG}}^{\text{max}2}$  can be adopted as an important figure-of-merit (FOM) of NLO polymers for the CWC application, with respect to SHG, on the condition of unignorable absorption coefficients. From Fig. 7, we can comprehend that the values of the FOM is higher than 0 dB, except only for the condition

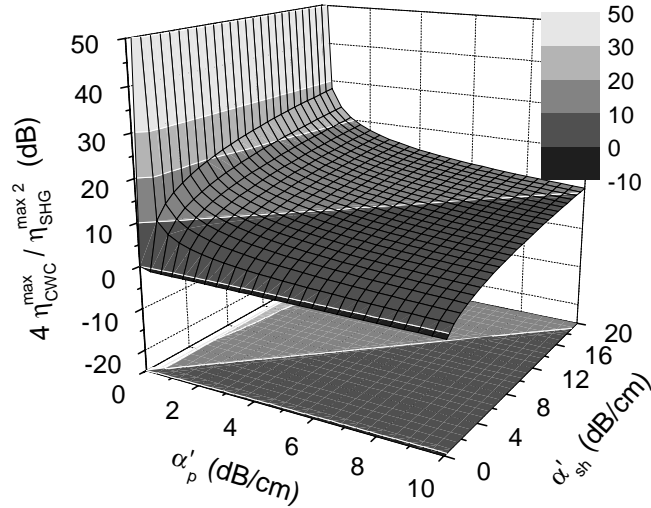


Fig. 7. Relational comparison between the maximum conversion efficiencies of the CWC and SHG processes, which can be used as a figure-of-merit of NLO polymers for the application to CWC.

of  $\alpha'_{sh}/\alpha'_p \approx 0$ , and exhibits a tendency to be enhanced more rapidly as  $\alpha'_{sh}/\alpha'_p$  increases.

### 3. Discussions

#### 3.1. Validity Criteria

Since all the above results are derived under the assumption given by Eq. (2), we should take care not to utilize them beyond their validity limit. Thus, we have to examine their validity condition, in the first place. Based on the above analytical approach and the results of numerical simulations with a system of equations more comprehensive than Eq. (1), we find the validity criteria expressed as

$$\eta_{\text{SHG}}(z) \ll \frac{c\epsilon_0 n_p^2 n_{sh} \alpha_p^2 \lambda_p^2}{32\pi^2 d_{\text{eff}}^2 I_p(0)} \quad (11)$$

and

$$z \leq z_{\text{SHG}}^{\text{max}} \quad (12)$$

Although we obtained an additional criterion given by

$$\eta_{\text{SHG}}(z) \cdot \eta_{\text{CWC}}(z) \ll \frac{c\epsilon_0 n_s n_c n_{sh} \alpha_s^2 \lambda_s^2}{32\pi^2 d_{\text{eff}}^2 I_p(0)} \cdot \exp(-\alpha_s z) \quad (13)$$

from Eq. (1), it looks a much looser condition compared to Eq. (11) in our situation, and hence, is not accepted.

One may notice that Eq. (11) looks very dubious because Eq. (11) connotes that the above results in section II are valid when the SH intensity, given by  $I_{sh}(z) = \eta_{\text{SHG}}(z)I_p(0)$ , is far below a constant value determined by material parameters and the pump wavelength, but unconcerned with a ratio to the pump intensity. However, it can be proved that Eq. (11) implies the condition of  $I_{sh}(z) \ll I_p(0)$  properly since it can be rewritten after a little algebra as

$$\eta_{\text{SHG}}(z) \ll \frac{1}{2} \alpha_p z \mathcal{L}_{\text{SHG}}^{1/2}(z) \quad (11')$$

and the relation of  $\alpha_p^2 z_{\text{SHG}}^{\text{max}^2} \mathcal{L}_{\text{SHG}}^{\text{max}} \leq 1$  is always satisfied according to the analytical results of Ref. [6]. It should be also noted that the validity conditions of Eqs. (11) and (12) are identical to those for SHG [6] and correspond to very tight conditions if we are interested only in the results for the CWC efficiency. Thus, the conditions can be much alleviated in such cases. However, it seems impossible to derive such alleviated criteria in a finite form because of the complicate contribution of the SH intensity to the CWC process.

Till now, we have considered only the second-order NLO interactions. In most cases where one deals with the cascaded second-order processes, one needs to consider the effects due to direct third-order processes as well. Actually several authors have reported that both effects often have comparable magnitudes and thus the third-order NLO effects cannot be simply neglected [21–23]. By looking over those reports, we can reach the conclusion that the effects of direct third- and cascaded second-order processes are comparable when a third-order process is phase-matched. In this situation, the two successive second-order processes, which contribute to the third-order effect, have their respective phase mismatches that countervail each other. Then, the effective third-order susceptibility due to the cascaded second-order processes is expressed by  $[\chi^{(2)}]^2/n\Delta n$ , where  $n$  is a refractive index and  $\Delta n$  is the index mismatch derived from the phase mismatch [22, 24, 25]. Considering the facts that  $\chi^{(2)}$  and  $\chi^{(3)}$  are roughly on the order of 1 pm/V and 100 pm<sup>2</sup>/V<sup>2</sup>, respectively, and that  $\Delta n$  is typically of order 10<sup>-2</sup>, we can certify the comparable magnitudes of direct third- and cascaded second-order effects.

However, in the situation of the CWC process, all of the relevant third- and second-order processes satisfy almost their PM conditions simultaneously. As the result, the effect of the cascaded second-order processes becomes predominant over that of the direct third-order process, for  $\Delta n$  approaches to zero. This fact is reflected in Eq. (4) by the dependence of  $z^4$ , while a typical second- or third-order phase-matched process is represented by the dependence of  $z^2$ . Therefore the neglect of a direct third-order NLO interaction in Eq. (1) can be justified well enough.

### 3.2. Estimation of the Maximum CWC Efficiency

#### 3.2.1. Case I

Till now, our group has developed a number of NLO polymers. Some of them exhibit very high NLO coefficients of  $d_{33} \simeq 60$  pm/V. These values correspond to the reported EO coefficients of  $r_{33} \simeq 32$  pm/V [10], which were attained by metal-contact electric poling at an applied electric field of 100 V/ $\mu\text{m}$ . The absorption coefficients of these polymers were measured as  $\alpha'_p \simeq 3.0$  dB/cm and  $\alpha'_{sh} \gtrsim 30$  dB/cm at the 1.55- $\mu\text{m}$  and 0.78- $\mu\text{m}$  wavelength regions, respectively. Since they were developed for EO applications [26–28], their absorption coefficients at the SH wavelength region were not carefully controlled.

With the material parameters given above, the value of the efficiency coefficient  $h_{\text{CWC}}^{\text{max}}$  is obtained to be not more than -127.1 dB at the optimum CWC interaction length  $z_{\text{CWC}}^{\text{max}} = 1.9$  cm, and the FOM  $4\eta_{\text{CWC}}^{\text{max}}/\eta_{\text{SHG}}^{\text{max}^2}$  for CWC is given by  $\sim 18.0$  dB. Provided that the QPM condition for the relevant SHG process can be satisfied by an ideal QPM structure composed of the periodic repetition of  $+d_{33}$  and  $-d_{33}$  with a duty ratio of 50%, the effective NLO coefficient is given by  $d_{\text{eff}} = 2d_{33}/\pi \simeq 38$  pm/V. Then, we can expect from Eq. (10') that  $\eta_{\text{CWC}}^{\text{max}} = 1.7\%$  (-17.8 dB) can be achieved at the pump intensity of  $I_p(0) = 200$  kW/cm<sup>2</sup> when  $I_s(0)$  is much less than  $I_p(0)$ . In our previous study [6], we reported that the maximum SHG conversion efficiency of  $\eta_{\text{SHG}}^{\text{max}} = 3.3\%$  (-14.8 dB) is estimated for the same polymer at the optimum SHG interaction length given by  $z_{\text{SHG}}^{\text{max}} = 0.58$  cm. It is noticeable that the value of  $\eta_{\text{CWC}}^{\text{max}}$  is much comparable to that of  $\eta_{\text{SHG}}^{\text{max}}$ .

Our direct numerical simulation with a system of equations generalized more than Eq. (1), which also includes terms related to phase mismatches, yields the maximum CWC efficiency

of  $\eta_{\text{CWC}}^{\text{max}} = 1.2\%$  at  $z_{\text{CWC}}^{\text{max}} = 1.7 \text{ cm}$  when  $I_p(0) = 200 \text{ kW/cm}^2$  and  $I_s(0) = 1 \text{ kW/cm}^2$ . We can find that there is a somewhat difference between the results of the analytical estimation and the numerical simulation. This is due to the effect of saturation in the SHG conversion efficiency at  $z > z_{\text{SHG}}^{\text{max}}$ , which corresponds to the violation of the validity criterion given by Eq. (12).

### 3.2.2. Case II

We developed another kind of NLO polymers as well, which exhibit much reduced absorption coefficients of  $\alpha'_p \simeq 1.8 \text{ dB/cm}$  and  $\alpha'_{sh} \simeq 3.8 \text{ dB/cm}$  at  $1.55 \mu\text{m}$  and  $0.78 \mu\text{m}$ , respectively. They also possess somewhat lowered NLO coefficients given by  $d_{33} \simeq 35 \text{ pm/V}$  [11] owing to the trade-off between the nonlinearity and absorption coefficients [5, 6].

For these polymers,  $h_{\text{CWC}}^{\text{max}}$  is obtained as  $-108.5 \text{ dB}$  at  $z_{\text{CWC}}^{\text{max}} = 4.5 \text{ cm}$  and the FOM for CWC is given by  $10.4 \text{ dB}$ . With an ideal QPM structure having  $d_{\text{eff}} \simeq 22 \text{ pm/V}$ , we can then achieve  $\eta_{\text{CWC}}^{\text{max}} = 13.9\%$  ( $-8.6 \text{ dB}$ ) at  $I_p(0) = 200 \text{ kW/cm}^2$  when  $I_s(0) \ll I_p(0)$ . This is also a much comparable value to  $\eta_{\text{SHG}}^{\text{max}} = 22.4\%$  ( $-6.5 \text{ dB}$ ) obtained at  $z_{\text{SHG}}^{\text{max}} = 2.35 \text{ cm}$  for the same polymer [6]. In the meantime, direct numerical simulation gives the result of  $\eta_{\text{CWC}}^{\text{max}} = 9.2\%$  at  $z_{\text{CWC}}^{\text{max}} = 4.2 \text{ cm}$  when  $I_p(0) = 200 \text{ kW/cm}^2$  and  $I_s(0) = 1 \text{ kW/cm}^2$ . The difference in  $\eta_{\text{CWC}}^{\text{max}}$  arises also from the saturation effect of the SHG and CWC efficiencies.

We can notice here that, despite of their lower NLO coefficients, these polymers present significantly higher CWC efficiencies than the former polymers in virtue of their low absorption coefficients at both the pump and SH wavelength regions. Moreover, if we take into account other side effects, such as photodegradation [8], related closely to the high absorption coefficients at  $0.78 \mu\text{m}$ , we can perceive that these polymers are rather suitable than the former ones for practical CWC devices.

### 3.2.3. Case III

The known 4-dimethylamino-4'-nitrostilbene (DANS) side-chain polymer can also give us an important guide related to the development and applications of NLO polymers for CWC. The absorption coefficients of this polymer at  $1.55 \mu\text{m}$  and  $0.78 \mu\text{m}$  are estimated as  $\alpha'_p \simeq 0.4 \text{ dB/cm}$  and  $\alpha'_{sh} \simeq 30 \text{ dB/cm}$ , respectively, and the value of the NLO coefficient is given by  $d_{33} \simeq 25 \text{ pm/V}$  [5].

From these parameters, we obtain  $h_{\text{CWC}}^{\text{max}} = -108.8 \text{ dB}$  at  $z_{\text{CWC}}^{\text{max}} = 12.2 \text{ cm}$ . Meanwhile, the FOM for CWC exhibits a very high value of  $30.5 \text{ dB}$  mainly due to the very low value of  $\alpha'_p$ . Then, an ideal QPM structure with  $d_{\text{eff}} \simeq 16 \text{ pm/V}$  is expected to yield  $\eta_{\text{CWC}}^{\text{max}} = 3.4\%$  ( $-14.7 \text{ dB}$ ) at  $I_p(0) = 200 \text{ kW/cm}^2$  and  $I_s(0) \ll I_p(0)$ , while the maximum SHG conversion efficiency is estimated as  $\eta_{\text{SHG}}^{\text{max}} = 1.1\%$  ( $-19.8 \text{ dB}$ ) at  $z_{\text{SHG}}^{\text{max}} = 1.08 \text{ cm}$  [6]. It is quite remarkable that the value of  $\eta_{\text{CWC}}^{\text{max}}$  is even higher than that of  $\eta_{\text{SHG}}^{\text{max}}$  for this polymer. This is due to the fact that  $\eta_{\text{CWC}}^{\text{max}}$  can be significantly increased relatively to  $\eta_{\text{SHG}}^{\text{max}}$  in the region of  $\alpha'_{sh}/\alpha'_p \gg 1$ , as shown in Fig. 7. Though, the increased value is still much lower than the value of  $\eta_{\text{CWC}}^{\text{max}}$  estimated for the case II even with a longer  $z_{\text{CWC}}^{\text{max}}$ .

From the results of our numerical simulation,  $\eta_{\text{CWC}}^{\text{max}} = 2.1\%$  is obtained at  $z_{\text{CWC}}^{\text{max}} = 10.3 \text{ cm}$  when  $I_p(0) = 200 \text{ kW/cm}^2$  and  $I_s(0) = 1 \text{ kW/cm}^2$ . Since  $z_{\text{CWC}}^{\text{max}}$  is greatly higher than  $z_{\text{SHG}}^{\text{max}}$  for this polymer, the saturation effect is considerable for the SHG conversion efficiency. Hence, a relatively high difference is generated between the estimated and simulated values of  $\eta_{\text{CWC}}^{\text{max}}$ .

### 3.3. Further Considerations

It is appreciated from Fig. 6 that a further improvement of  $\eta_{\text{CWC}}^{\text{max}}$  can be achieved by a decrease in  $\alpha'_p$  even though such material tuning may accompany a noticeable increase in  $\alpha'_{sh}$ . Thus NLO polymers adequate for CWC applications may have a somewhat higher absorption coefficient at the SH wavelength region. This relieves considerably the knotty restriction in the develop-

ment and application of NLO polymers, which is originated from the trade-off between low absorption at the SH wavelength and high nonlinearity [5, 6]. However, such promising NLO polymers must exhibit a sharp declination in the absorption spectrum to ensure a low absorption coefficient at the pump wavelength, as discussed in Refs. [6] and [11].

Till now, we have considered only ideal QPM conditions that are described by the effective NLO coefficients given by  $d_{\text{eff}} = 2d_{33}/\pi$ . However, it was already reported that such high values of  $d_{\text{eff}}$  cannot be obtained with usual periodic poling methods in non-ferroelectric media [15,29]. According to the results in Ref. [29], the maximum achievable value of  $d_{\text{eff}}$  is limited to  $\sim 0.34 \cdot 2d_{33}/\pi$ . Besides, the induced  $d_{33}$  values of practical QPM WC devices cannot reach to the maximum available values of the applied NLO polymers themselves. This is due to the fact that the poling electric field applied for practical polymeric QPM devices should be generally restricted to a value much lower than that applied for material characterization in order to obtain devices with good stability and low propagation losses [15, 16]. Hence it is requisite to develop a sophisticated device scheme or poling method to utilize the advantage of the high NLO coefficients of NLO polymer materials.

The periodic poling process also causes a significant problem of additional propagation losses, which cannot be ignored especially at the SH wavelength [15, 16]. These additional losses affect all the above analytical results as if the respective absorption coefficients were increased by such amounts. Thus a decrease in the CWC efficiency is unavoidable to some extent, though it may not so grave as in SHG. We already reported in our previous papers [13, 15, 16] that the increased propagation losses have a close relation with the surface deformation of fabricated QPM WC devices, and apprehended that the deformation is resulted from the geometry of the poling electrode and the poling field distribution [29]. Therefore we expect this problem to be fairly mitigated by avoiding the surface deformation, e.g., via introducing a rigid layer below the upper electrode layer.

Furthermore, we had better take into account the effect of the photodegradation due to the one- or two-photon absorption on the performance of polymeric NLO devices [7, 8]. However, a discussion on this subject is not dealt in this paper because it can be referred in our previous paper [6].

#### 4. Conclusion

We have investigated theoretically the effect of the absorption and optical nonlinearity of NLO polymers on the CWC process, and also applied the analytical results to estimate the CWC efficiencies for several kinds of NLO polymers. According to the results, we could comprehend that the high absorption coefficient at the SH wavelength of a typical NLO polymer does not affect significantly the CWC efficiency, and thus, it is possible to obtain a CWC efficiency that is comparable to or even higher than the expected SHG conversion efficiency. These facts demonstrate that the CWC process can be utilized effectively as a promising WC technique for wavelength-division multiplexed optical communication networks. We also discussed several important restrictions of practical polymeric QPM WC devices and proposed probable solutions applicable to the development of NLO polymers and QPM WC devices for CWC applications.

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