Available online at www.sciencedirect.com
SCIENCE DIRECT®

Optics Communications 236 (2004) 203–208

OPTICS COMMUNICATIONS

www.elsevier.com/locate/optcom

Optimization of second harmonic generation in non-linear film structure

G. Dyankov [a,*,1], Huai-Yi Chen [b], Shiuh Chao [c], Sien Chi [a]

[a] Institute of Electro-Optical Engineering, National Chiao Tung University, 1001 Ta Hsueh Road, Hsinchu 30056, Taiwan, ROC
[b] Department of Electronic Engineering, Huafan University, No.1 Huafan Road, Shih-Ting, Taipei County, Taiwan, ROC
[c] Institute of Photonics Technology, National Tsing Hua University, No. 101 Kuang Fu Road, Hsinchu, Taiwan, ROC

Received 10 November 2003; received in revised form 2 March 2004; accepted 2 March 2004

Abstract

The problem about optimization of second harmonic generation (SHG) by a laser beam in a non-linear medium with dimensions less than the beam size has not been solved yet. We present a theoretical method providing an optimization of SHG in the case of non-linear film non-waveguide structure. We have found that even in the case of perfect phase matching there is an optimum length for maximum non-linear interaction. The dependence of generation efficiency on different type of non-linear profiles has been established and related with experimental results.

PACS: 42.65.Ak; 52.35.Mw; 42.65.Ky; 42.79.Nv; 78.66.Aw

Keywords: Nonlinear phenomena; Frequency conversion; Harmonic generation; Nonlinear films; Thermally poled fused silica

1. Introduction

The second harmonic generation (SHG) phenomena were at the center of interest in the 1960s when the problem of interaction between light and non-linear media had been solved for plane wave [1,2] and for focused Gaussian beam in different approaches [3–6]. The basic results in [6] related to SHG optimizations are cited in many books and handbooks for non-linear optics. The noteworthy feature of all consideration [1–6] is that non-linear media are infinite.

In this paper, we solve the problem for SHG optimization in non-linear media with dimensions compatible and less than fundamental beam aperture with assumption of non-waveguide effects. The problem rose since non-linear properties in thin films (poled glasses, and poled fused silica) have been actively studied. In this paper, we propose a method enabling results valid in the case of infinite medium [6] to be applied for non-linear film (or strip) structure and permitting to find

* Corresponding author. Tel.: +86635712121x31584; fax: +86635716631.
E-mail address: dyankov@cc.nctu.edu.tw (G. Dyankov).
1 Present address: Institute of Solid State Physics, 72 Tzarigradsko Shousse Blvd., 1784 Sofia, Bulgaria.

0030-4018/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.optcom.2004.03.006

optimal structure length and fundamental beam spot size. The method treats the case of non-constant non-linear coefficient. The theoretical results are illustrated with experimental data [7,8].

2. Theory

For the materials (glasses or fused silica) we are interested in, the poling process primarily yields change in the non-linearity and only small or no change in the refractive index. Substrate is from the same material unaffected by poling process. In our consideration non-waveguide effects are assumed. The correctness of this assumption is obvious in the case of periodical poling–periodically modulated refractive index cannot crate waveguide. In the case of uniform poling asymmetric waveguide structure could be considered: cladding (air, with refractive index n_0), core (poled film – refractive index n_1) and substrate (refractive index n_s). This structure is not in waveguide regime when [9]:

$$ka\sqrt{n_1^2 - n_s^2} < 0.5\arctan((n_s^2 - n_0^2)/(n_1^2 - n_s^2)), \tag{1}$$

where k and $2a$ are wavenumber in a vacuum and waveguide width, respectively. This condition holds for fused silica, bearing in mind that poling depth (core width, respectively) is about 20 μm [8] and that poling process yields refractive index change below 9×10^{-5} [10]. The method proposed can be applied for uniform poling structure with parameters provided Eq. (1) is fulfilled.

Our method is based on the result reported in [6]. Written for the case of our interest (no absorption; focal position is in the center of structure) we claim:

$$P_2 = KP_1^2 lh(\Delta k, \beta, \xi). \tag{2}$$

Eq. (2) describes SHG exited by a Gaussian fundamental beam, where P_2, P_1, l represents, respectively, second harmonic power, fundamental power, and structure's length. Coefficients describing material optical properties are included in K. Variable $\xi \equiv z/b$ is a normalized distance of propagation in structure; $b \equiv k_1\omega_0^2$ is a confocal beam parameter (ω_0 – focal beam spot size); k_1 is fundamental wave number. The function $h(\Delta k, \beta, \xi)$ stands for the Gaussian fundamental beam intensity distribution and diffraction, phase mismatch Δk and double refraction effects ($\beta \equiv \alpha/b$, α is a double refraction angle). The last two factors are related to experimental conditions or are inherent in non-linear materials. Beam intensity distribution and diffraction are included in $h(\Delta k, \beta, \xi)$ integrated over the spot size. This integration can be changed in the case of film (or strip) media, since its dimensions of a cross-section perpendicular to a beam propagation direction are compatible or less than fundamental beam radius.

The proposed method extends the results of Eq. (2) for infinite non-linear media to the case when fundamental power is constrained by the medium dimensions. Fig. 1 illustrates focusing configuration and all structure dimensions. To extend the solution of Eq. (2) to our requirements, it is necessary to calculate what part of fundamental power is constricted in the film structure, namely it is essential to take the integral:

$$f_c(\xi) \equiv \int_{S(\xi)} I_1(x, y, \xi)\,\mathrm{d}x\,\mathrm{d}y, \tag{3}$$

where

$$I_1(x,y,\xi) = \left|(A/(1+\mathrm{i}\xi))\exp(-(x^2+y^2)/\omega_0^2(1+\mathrm{i}\xi))\right|^2 \tag{4}$$

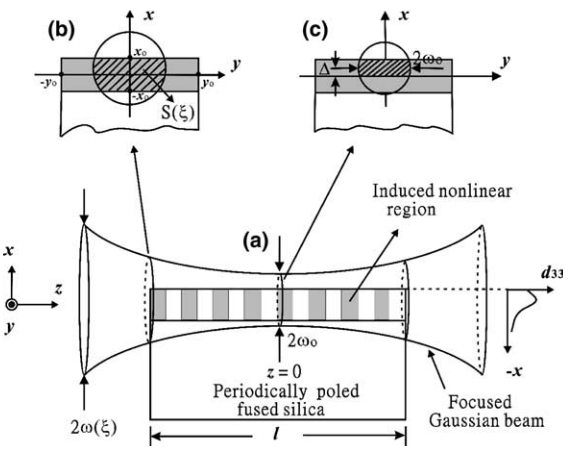

Fig. 1. (a) Focusing configuration in our consideration. (b and c) Characteristic dimensions in different sections of non-linear structure.

and $2x_0, 2y_0$ are film structure dimensions; A is fundamental beam amplitude; x and y hold for $x^2 + y^2 = \omega_0^2(1 + 2\xi)$ when $y_0 > \omega_0(1 + 2\xi)$. In fact, $S(\xi)$ is the overlap between Gaussian beam spot at distance ξ and cross-section of film structure, which defines an effective area of interaction (see Fig. 1(b)). In the case $S(\xi) = \pi\omega^2(\xi)$ we define:

$$f_c(\xi) \equiv 1. \tag{5}$$

Our approach provides mathematical insight: the function $f_c(\xi)$ plays a role of weighting coefficients accounting for the fundamental power captured by a film structure. Eq. (2) is extended to

$$P_2(\xi) = KP_1^2 f_c(\xi) l h(\Delta k, \beta, \xi). \tag{6}$$

In the case of perfect phase matching $\Delta k = 0$ and in the limit $\xi \ll 1$ as well as $\beta \to 0$, Eqs. (2) and (6) reduce to

$$P_2(l) = KP_1^2 l^2 / A \tag{7}$$

in which A is a focal spot area. Under the same assumptions ($\Delta k = 0, \xi \ll 1, \beta \to 0$) Eq. (7) can be applied to film structure section of length ξ when $A \equiv S(\xi)$ and then Eq. (7) takes a form:

$$P_2(\xi) = K f_c^2(\xi) \xi^2 / S(\xi). \tag{8}$$

The results from Eq. (8) coincide with the results of Eq. (6), what confirm our model. In Eq. (8) $f_c(\xi)$ is the fundamental power in film media and definition in Eq. (5) is not valid. The other quantities in Eq. (8) hold for definitions in Eqs. (3) and (4).

For the theoretical model in [6], non-linear coefficient d is constant and it is included in K coefficient. Our method permits to treat the case when non-linear coefficient is a function of coordinates, e.g., $d = g(x)$ which can be included in the integral of Eq. (3):

$$f_c^{\text{eff}}(\xi) \equiv \int_{S(\xi)} I_1(x, y, \xi) g^2(x)\, dx\, dy. \tag{9}$$

The quantity $f_c^{\text{eff}}(\xi)$, playing the role of effective fundamental power involved in SHG and in the case $d \neq$ constant, appears in Eq. (6) instead of $f_c(\xi)$; other quantities defined in Eq. (4) are the same.

3. Results and discussion

To illustrate our method we consider SHG in thermally poled fused silica: $n_\omega = 1.44963$, $n_{2\omega} = 1.46071$, $\lambda_\omega = 1.06$ μm [7]. Measurements performed by prism-coupler technique showed no waveguide mode in the uniformly poled silica. Transmission spectrum of the fused silica before and after thermal poling, also make no differences which implies the variation of refractive index through poling is consistent with estimation from Eq. (1) and with [10].

The results reported below relate to SHG power in periodically poled non-linear film structure where $\Delta k = 0, \beta = 0$ and $h(\Delta k, \beta, \xi) \equiv h(0, \xi)$. For the sake of convenience we define $P_2|_{S=\pi\omega^2(\xi)}$ $(\xi \to \infty) \equiv 1$ which is used for normalization.

Fig. 2 shows SHG power for different spot size ω_0 of the fundamental beam as a function of ξ for the case of film thickness $2x_0 = 40$ and 18 μm, respectively. For $\omega_0 = 1$ μm fundamental beam propagates entirely within the structure – the behavior is well known [1,6] for the case of infinite structure. Increasing ω_0 decreases SHG power – only a part of the fundamental beam is involved in

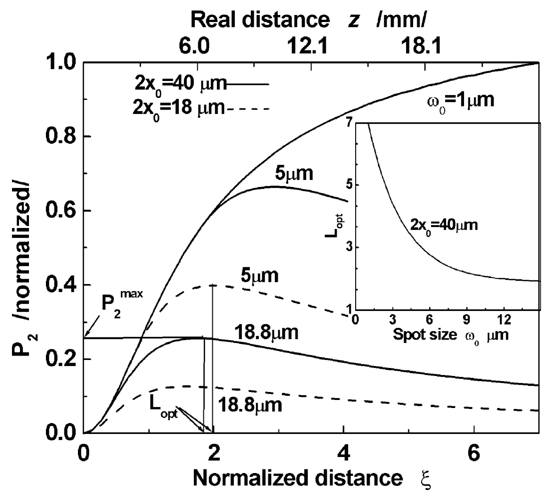

Fig. 2. The dependence of normalized SHG power on propagation distance for different beam spot size ω_0 and different film thickness $2x_0$; optimal length and optimal power are pointed. Inset: the dependence of optimal length on the spot size.

conversion process. Obviously, there is an optimum length when SHG power is maximal. Three important points provide physical insight into the optimum length and maximum SHG power: SHG occurs only in the film medium; film medium is not a waveguide – it cannot confine fundamental and SHG waves; the aperture of film structure limits the registration of SHG (in numerical simulations and in experiment). The optimal length ($L_{\rm opt}$) and maximum SHG power ($P_2^{\max}$) for $2x_0 = 40$ μm are also indicated in Fig. 2. For $\omega_0 = 18.8$ μm, $L_{\rm opt} = 1.82$ ($l_{\rm opt} = 5.5$ mm), $P_2^{\max} = 0.26$; for $\omega_0 = 5$ m, we have $L_{\rm opt} = 2.1$ ($l_{\rm opt} = 6.3$ mm) and $P_2^{\max} = 0.4$. The optimum length depends both on focal spot size and film thickness (see Fig. 2 – dashed curves), e.g., for $2x_0 = 18$ μm and $\omega_0 = 18.8$ μm, $L_{\rm opt} = 1.54$ ($l_{\rm opt} = 4.6$ mm) and $P_2^{\max} = 0.13$. Our numerical simulations have shown that $L_{\rm opt}$ and $P_2^{\max}$ decrease exponentially with spot size increasing (at fixed film thickness). The result for $L_{\rm opt}$ is illustrated at Fig. 2 inset. The second-order non-linearity of thermally poled fused silica is a function of depth [8,11]. In this case Eq. (9) gives the effective value of fundamental power involved in SHG. It was found [8] that non-linear profile correlates with buried Gaussian function $g(x) = d_{33}\exp(-(x-6.1/19.3)^2)$ and step-like profile ($d_{33} =$ constant). In [11] it was proposed that non-linear profile is an exponential decay function: $g(x) = d_{33}\exp(-(x_0 - x)/x_0)$. Different types of non-linear profile have considerable influence on SHG efficiency as illustrated in Fig. 3 (SHG is proportional to $lf_c^{\rm eff}(\xi)h(0,\xi)$ factor). Calculations are performed for periodically poled structure experimentally investigated in [7,8]: film thickness – 18 μm, length – 7 mm, $\omega_0 = 18.8$ μm. From experimentally measured SHG efficiency [7], non-linear buried Gaussian profile [8] and calculated value $lf_c^{\rm eff}(\xi)h(0,\xi) \cong 0.43$ (Fig. 3) it was obtained $d_{33}^{\rm eff} \approx 0.01$ pm/V for periodically poled structure. This coincides with estimation in [7], performed for the case of step-like profile, so it is over estimated with about 60% as indicated in the simulation results. The $d_{33}^{\rm eff}$ value includes the effect of structure constraints on the fundamental power and is about one order of magnitude less than that measured by Maker fringe method [7].

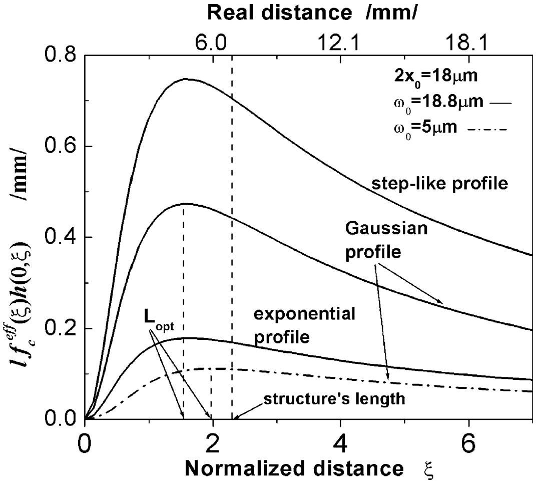

Fig. 3. Computed curves of $lf_c^{\rm eff}(\xi)h(0,\xi)$ for different non-linear profiles as a function of propagation distance; the length of sample $l = 7$ mm and $L_{\rm opt}$ are pointed.

In Fig. 3, we see that at different non-linear profiles $L_{\rm opt}$ keeps his value – $L_{\rm opt}$ is determined by the fundamental power involved in frequency conversion; non-linear profile only redistributes the power in the medium. As an illustration: $L_{\rm opt} = 1.98$ when $\omega_0 = 5$ μm and Gaussian profile (see dashed curve in Fig. 3) – the same in the case of step-like profile (see Fig. 2).

Calculation of $f_c^{\rm eff}(\xi)$ can be performed when laser spot scans transversely across the non-linear film (Fig. 1(c)). Fig. 4 shows SHG power as a function of displacement Δ for the same structure and non-linear profiles as in Fig. 3 and for focal spot size $\omega_0 = 1.5$ μm. Results suggest that the non-linear profile can be measured in a precise experiment by transversely spot scanning – P_2 dependence on Δ at different profiles gives unambiguous information.

The modified form of function $h(0,\xi)$ – Eq. (2.25) in [6] or more convenient form of Eq. (34) in [5] can be used in our method. (When Eq. (34) in [5] is used, should be aware of the non-correct ratio between double refraction α and sample length l – see critical remarks in [6]). The modified form of $h(0,\xi)$ permits to find the optimal focal spot size for film structure – an important procedure, because usually in experiment it is easier to

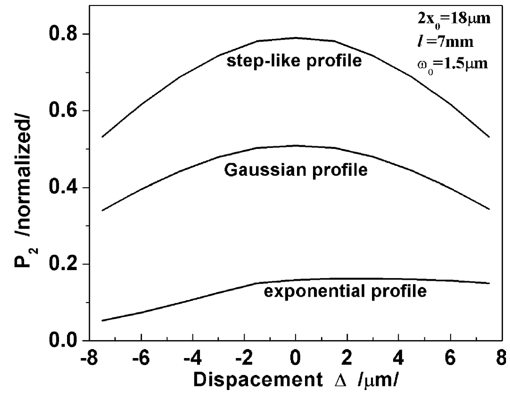

Fig. 4. Transverse spot scanning: dependence of normalized SHG power on displacement Δ for different non-linear profiles.

adjust the focal spot size rather than the sample length.

The result of spot size optimization for the film structure experimentally investigated in [7] ($l \neq L_{opt}$ – see Fig. 3) is shown in Fig. 5 for step-like and Gaussian non-linear profiles. Again, non-homogeneous non-linear profile plays an important role: Gaussian profile decreases SHG power with about 37% compared to step-like profile whereas the optimal spot size ω_0^{opt} keeps his value for different non-linear profiles. The upper curve corresponds for bulk sample ($2x_0 \gg \omega(\xi)$) made from the same material (non-linearity extends all through the sample) and is shown just for comparison. Our simulations have shown that optimal spot size for

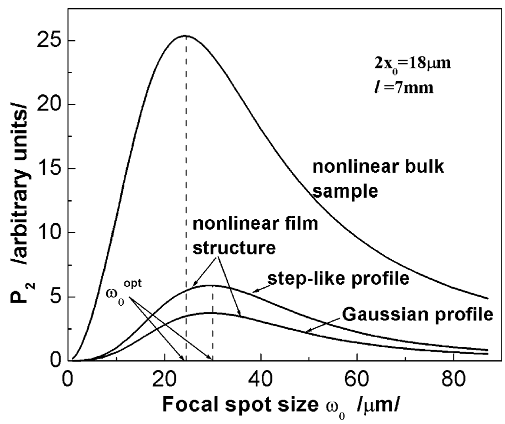

Fig. 5. Optimization of SHG efficiency with respect to the focal spot size ω_0 for structure experimentally investigated.

film structure is larger than that for bulk sample. So, we can propose a practical recipe: to begin the optimization at ω_0^{opt} for bulk sample (which is not difficult to calculate) and then increase ω_0 to search for maximum SHG power of non-linear film sample. It is clear (Fig. 5) that optimization with respect to ω_0 for a fixed profile is not a very critical adjustment. The sharpest in the peaks is within 10% of its maximum over the range $23<\omega_0<36$ μm and proposed recipe could be done easy.

4. Conclusions

The method we have proposed extends the results for optimization of SHG in infinite structure to the case of film (or slab) structure where non-waveguide effects take place. This method can be applied not only for the case of ordinary phase matching but also for optimal phase matching and double refraction. The method gives the possibility to optimize both – the structure length and fundamental focal size to achieve maximum SHG efficiency in non-linear film structure.

The approach of Eq. (8) can be useful in theory too. It is possible to include in Eq. (8) the diffraction involving the local waveform curvature and to solve the problem completely numerically even in the case $\Delta k \neq 0$. This approach is complicated in the case of double refraction. Moreover, Eq. (8) is correct only in weakly focusing approach, e.g., at $\omega_0 \geqslant 10$ μm for fused silica and $\lambda_\omega = 1.064$ μm. That's why we prefer the form of Eq. (6) using the well-approved results of [6].

In this paper, we have presented results on SHG in non-linear film non-waveguide structure in the case of ordinary phase matching $\Delta k = 0$. The main conclusion of our study is that the film structure has an optimum length with maximum generation efficiency even in the case of perfect phase matching. The optimum length and maximum SHG power decrease exponentially when fundamental spot size increases. The generation efficiency for structure with non-homogenous non-linear profile is considerably lower than efficiency of step – like profile: with about 37% for Gaussian profile and with about 75% for exponential profile. A procedure about SHG optimization, tuning fundamental spot size, has been suggested.

Acknowledgements

This work is supported by National Science Council (NSC) of the Republic of China under the Contract NSC 91-2215-E-007-025.

References

[1] J.A. Armstrong, N. Blombergen, J. Ducuing, P. Pershan, Phys. Rev. 127 (1962) 1918.
[2] D.A. Kleinman, Phys. Rev. 128 (1962) 1761.
[3] G.D. Boyd, A. Askin, J. Dziedzic, D.A. Kleinman, Phys. Rev. 137 (1965) A1305.
[4] D.A. Kleinman, A. Askin, G.D. Boyd, Phys. Rev. 145 (1966) 338.
[5] J.E. Bjorkholm, Phys. Rev. 142 (1966) 126.
[6] G.D. Boyd, D.A. Kleinman, J. Appl. Phys. 39 (1968) 3597.
[7] H.Y. Chen, J.S. Sue, Y.H. Lin, S. Chao, Opt. Lett. 28 (2003) 917.
[8] H.Y. Chen, J.S. Sue, Y.H. Lin, C.S. Tsai, P.T. Wu, S. Chao, J. Appl. Phys. 94 (2003) 1531.
[9] D. Marcuse, Theory of Dielectric Optical Waveguides, Academic press, New York, 1992.
[10] F.C. Garcia, L. Vogelaar, R. Kashyap, Opt. Exp. 11 (2003) 3041.
[11] R.A. Myers, N. Mukherjee, S.R.J. Brueck, Opt. Lett. 16 (1991) 1732.