Efficient one-third harmonic generation in highly Germania-doped fibers enhanced by pump attenuation

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We provide a comprehensive study on one-third harmonic generation (OTHG) in highly Germania-doped fiber (HGDF) by analyzing the phase matching conditions for the step index-profile and optimizing the design parameters. For stimulated OTHG in HGDF, the process can be enhanced by fiber attenuation at the pump wavelength which dynamically compensates the accumulated phase-mismatch along the fiber. With 500 W pump and 35 W seed power, simulation results show that a 31% conversion efficiency, which is 4 times higher than the lossless OTHG process, can be achieved in 34 m of HGDF with 90 mol. % GeO₂ doping in the core.

1. Introduction

Although third harmonic generation (THG) has been intensively investigated both theoretically and experimentally in past literature [1–4], the inverse process of one-third harmonic generation (OTHG) remains a new topic that has been attracting increasing research attentions. Previous studies on OTHG categorize the process into two kinds, namely, spontaneous downconversion and stimulated downconversion. The former was recently analyzed by Gravier [5], Corona [6] and Richard [7]. Inherently, however, the conversion efficiency is extremely low. Stimulated downconversion, on the other hand, is more appealing as the OTHG process can be seeded to achieve high conversion efficiency. In [8], Afshar et al. report a method for evaluating the maximum conversion efficiency of lossless THG and OTHG process, in which the maximum conversion efficiency is determined by waveguide structure and total input power. However, in real experiments, attenuation always exists and affects the total power along the interaction medium. Furthermore, because of the triple relationship of wavelength between pump and signal in OTHG, the loss profile at the two wavelengths can be quite different, which should consequently be taken into consideration along with the waveguide structure and power level when evaluating the performance of OTHG. High conversion efficiency can be expected by optimizing these parameters.
Highly Germania-doped fiber (HGDF) consisting of a Germania-Silica core within a Silica cladding has attracted extensive interest in the past years [9–13]. Also, HGDF is a good candidate for OTHG because of its capability to achieve inter-modal phase matching and low attenuation characteristics around 0.5 µm and 1.5 µm. However, to date, the role of doping concentration in HGDFs for OTHG has yet to be addressed in detail.

In this paper, a comprehensive study of the stimulated OTHG process in HGDFs is performed. The phase matching parameters in step-index fiber are designed. Different modes are discussed in terms of overlap integrals and the most suitable one is then chosen for inter-modal phase matching. The performances of different HGDFs are simulated and evaluated along with the possibility for pump attenuation in playing a contributing role towards efficient OTHG.

2. Phase matching in HGDFs

OTHG and THG share the same phase matching conditions, which have previously been discussed for glass microfibers [1, 2]. The main disadvantage is that the interactive length is limited to several centimeters because of the difficulty in fabricating long microfibers whilst maintaining the small diameter required for phase matching. High-delta micro-structured fibers were also employed to realize phase matching [14]. More recently, Tarnowski et al. proposed an approach to compensate for the phase mismatch for THG by using quasi-phase matching technique based on the grating written in the fiber [15]. However, the fabrication processes of these fibers are quite complicated. On the other hand, high numerical aperture (NA) fibers offer an attractive choice to achieve phase matching because the big fibers can be straightforwardly fabricated to much longer interactive lengths [6, 16]. Here we demonstrate a detailed analysis for phase matching conditions in HGDFs.

In the process of OTHG in optical fibers supporting several modes, a pump photon, oscillating at the circular frequency \( \omega_3 \) with wave vector \( k_3 \), splits into a set of three photons with circular frequencies \( \omega_1 = \omega_3/3 \) and wave vector \( k_1 = k_3/3 \). This condition is equivalent to:

\[
\text{n}_{\text{eff}}(\omega) = \text{n}_{\text{eff}}(3\omega),
\]

where \( \text{n}_{\text{eff}} \) is the effective index of the relevant mode. Here, we consider the phase matching between \( \lambda_3 = 0.532 \) µm (the pump wavelength) and \( \lambda_1 = 1.596 \) µm (the one-third harmonic wavelength). Figure 1 shows the material dispersion of pure and 10 mol. % GeO₂ doped silica. The Sellmeier functions for material dispersion are obtained from [17]. Due to uneven material dispersion curves, the cladding index at the pump wavelength exceeds the core index at the one-third harmonic wavelength. Hence, there is no overlap region between the effective index ranges for the two wavelengths in a step-index fiber with a core doping concentration of 10 mol. % and so phase matching between guided modes is impossible in such a weakly guiding fiber. In this case, guided modes at 1.596 µm can only match to the radiation modes at 0.532 µm, which suffer from large loss and small overlap integrals. The solution is to further increase the doping concentration to achieve a higher NA so that the effective index areas at the two wavelengths can overlap leading to the possibility of phase matching between two guided modes.
Fig. 1. Material dispersion of pure silica (cladding) and 10 mol. % GeO₂ doped silica (core). The arrows mark the range in which the effective indices of guided modes at $\lambda = 0.532 \, \mu \text{m}$ and $1.596 \, \mu \text{m}$ reside.

By solving the rigorous modal eigenvalue equations for a step index profile in HGDF [18], Fig. 2 demonstrates the effective indices of the modes at two wavelengths as a function of core diameter with a higher 30 mol. % doping concentration in the fiber core. Only hybrid HE and EH modes are considered since the overlap with transverse TE and TM modes is zero. Obviously, with sufficient doping concentration, a large number of high-order modes at $0.532 \, \mu \text{m}$ can find a core diameter for phase matching the mode HE₁₁ at $1.596 \, \mu \text{m}$. Because high-order modes are more difficult excite and have a smaller overlap with the fundamental mode, we restrict our discussion to the lower-order modes (EH₃₁, EH₁₂, HE₃₂, and HE₁₃).
Fig. 2. Effective indices of different modes as a function of core diameter with a Germania core doping concentration of 30 mol. %.

To assess which of the possible phase match points would be most efficient for OTHG, the modal overlaps $J_3$ between the HE$_{11}$(ω) and each higher order ω$_3$ mode were calculated according [1]:

$$ J_3 = \iint_{A_m} \left( F_1^* \cdot F_3 \right) \left( F_1^* \cdot F_3 \right) dS $$

where $F_1$ and $F_3$ denote the power-normalized electric field distributions of the HE$_{11}$ mode at 1.596 µm and the phase matched higher order mode at 0.532 µm, respectively. The characteristics of the four phase-matched modes mentioned above as a function of doping concentration are demonstrated in Fig. 3. It can be seen from Fig. 3(a) that their effective indices increase with doping concentration because of the higher refractive index of GeO$_2$. In Fig. 3(b), the phase matching diameters decrease with increased doping since the modes are more tightly confined within the core region. Although the diameter for phase matching with mode the HE$_{13}$ mode is larger than the others, this mode has the largest overlap integrals $J_3$, and is therefore the most promising choice for ensuring an efficient OTHG process. This finding is valid because both HE$_{11}$ and HE$_{13}$ are circularly symmetric and have their largest field intensity in the centre. Note that the overlap increases with increasing doping concentration as shown in Fig. 3(c), again due to the increase in modal confinement.
Fig. 3. (a) Effective index, (b) core diameter and (c) $J_3$ as a function of doping concentration for modes at 4 different phase matched points.

3. Analysis and simulation results

Assuming the fields at $\omega_1$ and $3\omega_1$ are in different but single propagating modes of the fiber, the fields can be described by slowly varying amplitudes $A_i(z)$, where $i = 1$ refers to the one-third harmonic, and $i = 3$ refers to the pump. The following set of equations can then be derived [1]:

\[
\begin{align*}
\frac{\partial A_1}{\partial z} &= -\frac{\alpha_i}{2} A_1 + i \gamma_0 [J_1 (A_1^2 + 2J_2 |A_1|A_2)A_1 + J_3 (A_3^2)A_1 e^{i\Delta\beta z}] \\
\frac{\partial A_3}{\partial z} &= -\frac{\alpha_i}{2} A_3 + i \gamma_0 [(6J_2 |A_1|^2 + 3J_3 |A_3| A_3)A_3 + J_3 A_1^2 e^{-i\Delta\beta z}] 
\end{align*}
\]

where $J_1$, $J_3$, $J_5$, and $J_6$ denote the modal overlap integrals for one-third harmonic self-phase modulation (SPM), pump harmonic cross-phase modulation (XPM), pump harmonic conversion, and pump SPM, $\alpha_i$ is the loss coefficient, $\Delta\beta = \beta(3\omega) - 3\beta(\omega)$ is the propagation constant mismatch, and $\gamma_0 = 2\pi n^{(2)}/\lambda$, where $n^{(2)}$ is the nonlinear refractive index coefficient. For GeO$_2$-doped silica, a linear relation with silica of $n^{(2)} = (0.0552\text{Conc} + 2.44) \times 10^{-20}$ m$^2$/W is used to estimate nonlinear refractive index, where Conc is the mole concentration [19]. The power of each mode is given by $|A_i|^2$. Defining $A_1 = \sqrt{P_1 e^{i\phi_1}}$, $A_3 = \sqrt{P_3 e^{i\phi_3}}$, and $\psi = \Delta\beta z + \varphi_3 - 3\varphi_1$, the coupled mode equations can be rewritten as:

\[
\begin{align*}
\frac{dP_1}{dz} &= -\alpha_i P_1 - 2J_2 \gamma_0 P_1^3 \frac{1}{P_3^2} \sin \psi \\
\frac{dP_3}{dz} &= -\alpha_i P_3 + 2J_2 \gamma_0 P_3^3 \frac{1}{P_3^2} \sin \psi
\end{align*}
\]
As shown in Eqs. (4) and (5), the power flow direction is determined by the sign of \( \sin \psi \), if \( \sin \psi > 0 \), it is THG process, otherwise OTHG. If \( \sin \psi < 0 \) is satisfied at the input of the interactive media and the right hand side of Eq. (6) is kept approximately to be 0, power can be transferred from the pump \( (P_3) \) to signal \( (P_1) \) continuously. However, as \( P_1 \) increases and \( P_3 \) decreases, Eq. (6) becomes non-zero and \( \psi \) deviates from \( \sin \psi < 0 \) because of the fiber loss and different Kerr coefficients \( (6J_2-3J_1) \) and \( (3J_5-6J_2) \), hence changing the varying rate and direction of the power flow. Additionally, according to Eq. (4), it is easy to note that the varying rate of \( P_1 \) can be quite low with a small seed light \( (P_1) \) at the input of the interactive media because the varying rate is proportional to \( P_1^{1.5} \). With small \( P_1 \), simply increasing the pump power contributes little to the conversion rate improvement. This feature is totally different with THG, whose power changing rate can be increased by strong pump, as shown in Eq. (5). Therefore, in order to obtain efficient OTHG, a potential method is to stimulate the process with powerful seed. The theoretical analysis for lossless OTHG proposed in [8] points out that the maximum conversion efficiency is dominated by the total input power and waveguide structure. In the above mentioned HGDF, according to our calculation, the relationship of overlap integrals cannot fulfill the special requirements mentioned in [8] for efficient OTHG process. However, because of the existence of loss which can be quite different at pump and signal wavelengths, the conversion mechanics are slightly different. Since the losses of pure silica and GeO₂ at 0.532 µm are about 11 dB/km and 300 dB/km and 0.2 dB/km and 0.6 dB/km at 1.596 µm [20], we use a linear relation to approximate the HGDF loss at different doping concentration, the attenuation coefficient at longer wavelength is much smaller which raises the possibility for dynamically compensating for the phase mismatch during propagation along fiber.

In order to investigate the parametric process in the above mentioned HGDF, we return to Eq. (6) which shows that \( \psi \) is determined by four terms including a linear \( \Delta \beta \) term (regarded as constant along a longitudinally invariant fiber). We also define the Kerr coefficients \( K_1 = 6J_2-3J_1 \), and \( K_3 = 3J_5-6J_2 \), whose values as a function of doping concentration are demonstrated in Fig. 4. Their behavior can be divided into three regimes:

![Fig. 4. \( K_1 \) and \( K_3 \) as a function of doping concentration](image-url)
(1) $0 < J_3 < K_3 < K_1$ (90 mol. % and 100 mol. % doping)

Since $K_1$ and $K_3$ are much larger than $J_3$, the last term of Eq. (6) can be neglected and the equation approximated as:

$$\frac{d\psi}{dz} = \Delta \beta + \gamma_0 (K_1 P_1 + K_3 P_3)$$

(7)

In this case, if $\sin \psi < 0$ is fulfilled at the input of the fiber, power can transfer from $P_3$ to $P_1$ due to OTHG process. Given this is a lossless process, the increasing rate of $K_1 P_1$ must be faster than the decreasing rate of $K_3 P_3$ because $K_1 > K_3$, and therefore after propagating along a short distance of fiber, the power will transfer from $P_1$ to $P_3$ again then oscillating between the two waves. However, the condition will be different considering large pump loss. Combining the effect of power attenuation caused by parametric processes (i.e. the transfer of power to the other wavelength) and fiber loss, the increment of $K_1 P_1$ can be comparable with the decrement of $K_3 P_3$ so that $\psi$ changes slowly and $P_1$ can build up continuously over a long fiber length. When $\psi$ changes to make $\sin \psi > 0$, the THG process starts and power begins to transfer from $P_1$ to $P_3$, reducing $K_1 P_1$ and increasing $K_3 P_3$. Similarly, the changing rate of $K_1 P_1$ is faster than $K_3 P_3$, meanwhile a large $\alpha_3$ prevents the increasing of $K_3 P_3$. These two terms cannot balance with each other, and therefore $\psi$ changes fast and it is difficult to maintain $\sin \psi > 0$. $\psi$ will vary such that $\sin \psi$ falls below 0 after a short propagation length and return to OTHG process again, as a result $P_1$ can accumulate. In this case, OTHG can benefit from the large loss at pump wavelength.

(2) $0 < J_3 = K_3 << K_1$ (40 mol. % and 50 mol. % doping)

This case is similar to the first one, but the balance between phase terms is more difficult because of the small values of $K_3$ and $J_3$. If the pump loss is too small, the $K_1 P_1$ term will dominate the phase varying rate and make the process close to the lossless one. Even if, the loss is large enough to compensate phase changing at the input of fiber, OTHG cannot benefit from pump attenuation for long distances since the pump power decays exponentially along the fiber. After a certain distance, the balance can be break.

(3) $K_3 < 0 < J_3 < K_1$ (30 mol. %, 60 mol. %, 70 mol. % and 80 mol. % doping)

In the OTHG process, both $K_1 P_1$ and $K_3 P_3$ increase while the changing direction of the $J_3$ term (the last term in Eq. (6)) is determined by the sign of $\cos \psi$ which can be positive or negative during OTHG process. Nevertheless, $J_3$ is too small compared with $K_1$, so $K_1 P_1$ and $K_3 P_3$ lead to a fast phase variation. While the OTHG buildup region is restricted to $-\pi < \psi < 0$, $\psi$ cannot stay in this favorable region for a long propagating length. Therefore, it is difficult to realize efficient OTHG.
To verify our analysis mentioned above, we used the Runge-Kutta method in MATLAB to numerically calculate Eqs. (2) and (3) to investigate the designed fiber with different doping concentrations. The pump and signal powers are set to be 500 W, and 35 W, respectively and initial phase difference $\psi(0) = -\pi/2$. For a lossy interaction, the conversion efficiency is given by $\eta = (P_{1out} - P_1(0))/P_3(0)$, where $P_{1out}$ is the power of the one-third harmonic at the output. In Fig. 5(a), we give the efficiency contour map with a 90 mol. % doping concentration with different $\Delta\beta$ values which make the right hand side of Eq. (5) approximately zero. In this case, an OTHG conversion efficiency of more than 30% can be achieved in a propagation constant mismatch range from $-51$ m$^{-1}$ to $-51.6$ m$^{-1}$ by taking advantage of the better balance between power transfer and power attenuation. It can be seen from Fig. 5(b) that within 34 m of fiber, the transfer of power from the pump to the seed (i.e. $-\pi < \psi < 0$) can continue over a net OTHG interaction length longer than that of THG. Although the system oscillates between OTHG and THG during propagation, during the latter process the phase change rate is much faster, and hence the phase quickly returns to the range required for OTHG. Consequently, the one-third harmonic experiences an overall growth, especially at the beginning of the fiber where the strong power decay of $P_3$ compensates the faster phase changing term $K_1P_1$. The OTHG power oscillates up until the pump depletes. Figure 5(c) illustrates the phase evolution along the fiber by solving Eq. (5), as predicted, $K_1P_1$ and $K_3P_3$ dominate the phase variation and balance with each other within 34 m fiber length. The total phase is more confined in the region favoring OTHG ($-\pi, 0$) as shown in the zoomed-in inset. After this length, the phase starts to change fast because the pump depletes and the nonlinear phase shift caused by the smaller $P_1$ is not sufficient to compensate $\Delta\beta$. In comparison, for lossless OTHG, the maximum conversion efficiency we can obtain is about 6% as shown in Fig. 5(d), which is 4 times smaller than the loss assisted process. Note that high conversion is not limited by the specific input power condition used here. For a 500 W pump, which can be easily achieved by frequency doubling...
from an Nd: YAG laser, 35 W is close to the minimum power needed for a high conversion (higher than 30%). To further investigate the efficiency with different input condition, Fig. 6(a) presents the maximum achievable efficiency and corresponding fiber length with 90 mol. % doping with respect to the seed power, when the pump power is fixed to be 500 W. Clearly, we can conclude that higher seed power leads to higher conversion efficiency and reduces the required fiber length. On the other hand, if the seed power is fixed at 35 W, the proposed scheme can also remain efficient when the input pump power varies in a large range as shown in Fig. 6(b). According to the main trends of the two curves, increasing the pump power will reduce the maximum conversion and increase the required fiber length. This is reasonable because a larger pump power speeds up the phase variation and contributes less to the power transferring rate comparing with seed power. However, since $0 < J_3 << K_3 < K_1$, the OTHG process is still overall enhanced over the fiber link. Therefore, in the specially-designed HGDF, efficient OTHG can be realized within a large power range of input pump and seed. Typically, they are not limited to the fixed 500 W pump and 35 W seed.

![Fig. 6. Maximum conversion and corresponding fiber length with respect to (a) seed power when pump power is fixed at 500 W and (b) pump power when seed power is fixed at 35 W in 90 mol. % doping HGDF.](image)

It should be noted that with high power input, Brillouin and Raman scattering process may act as additional attenuation mechanisms and affect the conversion efficiency. However, because there is no cavity or seed power at Brillouin and Raman scattering wavelengths in the proposed scheme, the power generated from these two nonlinear processes cannot be resonantly enhanced and must start from spontaneous processes with low efficiency. As the HGDF is designed to phase-match the pump and one-third harmonic wavelengths, with proper seed power, stimulated OTHG can happen immediately at the input of the fiber. Combining parametric attenuation and fiber loss, the pump power will deplete quickly and make other scattering processes more difficult to be efficient. Also, as shown in Fig. 6(b), a high efficiency is still expected even with lower pump powers and shorter fibers. For example, if the pump power is reduced to 100 W, nearly 35% conversion can still be achieved with 21 m of HGDF.
Fig. 7. (a) Contour map of conversion efficiency in 40 mol. % HGDF with different $\Delta \beta$, (b) conversion efficiency variation along fiber length with $\Delta \beta = -4.8 \text{ m}^{-1}$, and (c) phase variation along fiber.

For the case of a weaker 40 mol. % doping concentration, Fig. 7(a) shows a contour map of OTHG conversion efficiency long fiber length with different propagation phase mismatch $\Delta \beta$. Relatively strong OTHG happens in the region $-5 \text{ m}^{-1} < \Delta \beta < -4 \text{ m}^{-1}$, however, maximum conversion efficiency is less than 6%. Note that the regions of negative $\eta$ at $\Delta \beta > -3.5 \text{ m}^{-1}$ correspond to regimes in which the power is being transferred away from the seed to the pump wavelength. Figure 7(b) demonstrates the efficiency evolution with optimized $\Delta \beta = -4.8 \text{ m}^{-1}$. According to our analysis above, $\eta$ starts to build up at the input of the fiber reinforced by pump loss. Unfortunately, $\eta$ can no longer continue increasing after passing through the third peak because $P_3$, which decays exponentially along the fiber, cannot balance the huge phase changing caused by $K_1P_1$ which limits the buildup of $\eta$. The phase variation given in Fig. 7(c) shows the net interactive fiber length for OTHG and THG is nearly the same, causing the one-third harmonic power difficult to be accumulated. This can also be explained by the huge difference of $J_3$, $K_3$, and $K_1$. Power attenuation of $P_3$ caused by both fiber and parametric attenuation are not enough to slow down the phase variation.

As we mentioned above, with doping concentrations of 30 mol. %, 60 mol. %, 70 mol. % and 80 mol. %, pump loss is no longer favorable for OTHG. The performances of these four doping concentration are demonstrated in Figs. 8(a)–8(d), respectively. From the efficiency contour maps, it is apparent that $\eta$ cannot go up higher after reaching their first efficiency peak which limits conversion to $< 5\%$. 
4. Conclusion

In summary, we have designed step-index HGDFs for phase-matched OTHG and analyzed the performances for different fibers. Parameters such as effective indices, core diameters, and overlap integrals for HGDFs with different doping concentrations were investigated. In specially designed HGDF with core GeO₂ doping levels of 90 mol. %, the pump loss is no longer a disadvantage and can be used to compensate the accumulated phase-mismatch dynamically along the fiber for efficient OTHG. According to our simulation, 31% conversion efficiency which is nearly 4 times higher than the equivalent lossless process is achievable over 34 m of fiber with a doping concentration of 90 mol. % in the core. The results demonstrate that HGDFs can potentially be applied for efficient OTHG which is significant in the area of triple photons generation and parametric amplification.

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