Harmonic Generation via $\chi^3$ Intermodal Phase Matching in Microfibers

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Intermodally phase matched up- and down-conversion processes based on the third order nonlinearity have been proposed to efficiently generate light in the UV and mid-IR wavelength regions in solid core silica optical fibers and optical microfibers. We study waveguide parameters and practical considerations required for optimum conversion.

Keywords: Non Linear Fibers, fiber optics, frequency conversion, Wavelength Conversion, Multiwavelength

1. Introduction

Centrosymmetric amorphous media has been a candidate for the exploitation of third order $\chi^{(3)}$ nonlinearity for parametric nonlinear harmonic conversion processes. These processes include third harmonic generation (THG) and three-photon generation (TPG) which generate light at triple or one third of the pump frequency, respectively. The main requirement for these two processes is that the effective refractive indices ($n_{\text{eff}}$) of the pump ($\omega$) and harmonic ($3\omega$) frequencies are equal ($n_{\text{eff}}(\omega) = n_{\text{eff}}(3\omega)$). This so-called phase matching condition, however, cannot be satisfied for the fundamental modes of both frequencies, and the resulting phase mismatch due to material and waveguide dispersion leads to exceedingly low efficiencies. The same cannot be said about higher order modes, which generally have lower effective indices than the fundamental mode [1]. Therefore, intermodal phase matching has been proposed as a means to achieve high conversion efficiency in a number of high contrast waveguides, including microstructured fibers [2], optical fiber nanowires [3-6] and high numerical aperture (NA) fibers [7]. Optical microfibers provide an additional advantage, as the effective nonlinearity is strongly enhanced in such waveguides due to tight modal confinement [8]. In this paper,
the generation of light in UV and mid-IR via THG and TPG in optical microfibers are discussed theoretically to investigate the feasibility for efficient frequency conversion.

2. Material Consideration

Optical fibers have long been regarded to be opaque in the UV and mid-IR regions for two main reasons: (1) the dopants used to increase the core refractive index (primarily germanium and phosphorous oxides) had a strong absorption for wavelengths below 350 nm, and (2) there is a relatively high cumulative absorption if long lengths of fibers were considered [1,2]. Therefore, these regions are thought to be unsuitable for light generation. However, as we are interested in this region, we shall first consider the lowest possible losses present in silica in these wavelength regions.

Figure 1. Loss of pure silica in the UV until mid-IR [9-13]

The absorption of pure silica is presented in Figure 1 for the UV and mid-IR wavelength regions, respectively. Losses are smaller than 1 dB/cm in the wavelength range $0.2 < \lambda < 3.5 \mu m$, and exceeds 1 dB/mm only at $\lambda > 4.1 \mu m$. Therefore, pure
silica fibers (such as the submarine telecom fibers or in optical microfibers) can be employed to achieve light generation and propagation in the UV or mid-IR over lengths of the order of several mm without incurring excessive losses.

3. $\chi^{(3)}$ Processes

The phase matching condition required for THG and TPG can be achieved by tailoring waveguide geometry and index contrast, easily achieved by solving the rigorous modal eigenvalue equations assuming a step index profile for the waveguide [14]. The equations are solved for different modes for a range of microfiber diameters, with the phase matching diameter being the diameter at which the effective indices of the fundamental mode is the same as one of the harmonic modes. Following this, the conversion efficiency $\eta$ can be obtained by evaluating the amplitudes of both the fundamental and harmonic wavelengths, attained by solving the following coupled equations [2]:

$$\frac{\partial A_1}{\partial z} = i\gamma_0 [(J_1 |A_1|^2 + J_2 |A_3|^2)A_1 + J_3 (A_1^*)^2 A_3 e^{i\Delta \beta z} - \alpha_1 A_1]$$ (1)

$$\frac{\partial A_3}{\partial z} = i\gamma_0 [(6J_2 |A_1|^2 + 3J_5 |A_3|^2)A_3 + J_3 (A_{P1}^*)^3 e^{-i \Delta \beta z} - \alpha_3 A_3]$$ (2)

where the subscripts 1 and 3 refer to pump and harmonic signals, $A_1$ and $A_3$ are the amplitudes of the pump and the third harmonic mode, $J_i$ are the modal overlaps between different modes of the power-normalized electric field distributions, $\Delta \beta = \beta(3\omega) - 3\beta(\omega)$ is the detuning, $\gamma_0 = 2\pi n^{(2)}/\lambda$, $n^{(2)}$ is the nonlinear refractive index and $\alpha_1, \alpha_3$ are the losses in the waveguide.

The mathematical forms of the $J_i$ and their respective physical interpretations are given in Table 1, where we have denoted $F_i$ as the transverse electric mode field of either pump ($F_1$) or harmonic ($F_3$) [3].
Table 1. Physical interpretation of \( J_n \) integrals

<table>
<thead>
<tr>
<th>( J_n )</th>
<th>Mathematical expression</th>
<th>Physical interpretation</th>
</tr>
</thead>
<tbody>
<tr>
<td>( J_1 )</td>
<td>( \frac{1}{3} \iint_{\mathcal{A}_{NL}} (2</td>
<td>F_1</td>
</tr>
<tr>
<td>( J_2 )</td>
<td>( \frac{1}{3} \iint_{\mathcal{A}_{NL}} (</td>
<td>F_1</td>
</tr>
<tr>
<td>( J_3 )</td>
<td>( \iint_{\mathcal{A}_{NL}} (F_1^* \cdot F_3)(F_1^* \cdot F_1^*) dS )</td>
<td>Phase matched intermodal energy transfer via THG</td>
</tr>
<tr>
<td>( J_5 )</td>
<td>( \frac{1}{3} \iint_{\mathcal{A}_{NL}} (2</td>
<td>F_3</td>
</tr>
</tbody>
</table>

It should be noted that though the two equations are not symmetric, they do allow for the transfer to occur in both directions, i.e. from the pump to the harmonic wavelength as well as from the harmonic to the pump wavelength. The main difference is that the former case, THG can occur with just the presence of a strong long wavelength pump signal, whereas the latter case, TPG, will only occur with the simultaneous presence of a long-wavelength seed and a short-wavelength ‘pump’[15].

3.1 THG UV Generation

3.1.1 Theoretical considerations

As mentioned previously, THG UV generation in microfibers require phase matching between the fundamental and harmonic modes of the two wavelengths. The phase matching conditions for the different modes are shown in Figure 2, with the values given
in Table 2. Note that the pump wavelength employed is in the near-IR ($\lambda_1 = 0.8 \mu m$) such that the harmonic is generated in the UV.

![Graph showing effective index vs core diameter](image)

Figure 2. Dependence of the effective index with diameter for THG phase matching for UV generation for a fluorine doped silica microfiber in air. The pump wavelength, $\lambda_p$, is $0.8 \mu m$, with harmonic wavelength, $\lambda_h$, is $0.266 \mu m$.

The efficiency of the THG process to each mode is obtained by simultaneously solving equations 1 and 2. The waveguide is assumed lossless at the pump wavelength ($\alpha_1 = 0$), as there is negligible absorption in relatively short fibers [11], [16], and adiabatic microfibers have very small loss in the near infrared [17]. From equations 1 and 2, the main contributor to the THG is the $J_3$ overlap, as it is the modal overlap between the fundamental mode of the pump and the harmonic mode of the generated wavelength. The efficiency of each mode for wavelengths of $0.8 \mu m$ and $1.064 \mu m$ is given in figure 3, and it is clear that the highest overlap is between the fundamental $HE_{11}$ mode and the $HE_{12}$ mode, followed by the $HE_{31}$ mode. Furthermore, from Figure 4, the value of $J_3$ increases for decreasing wavelengths, indicating that the nonlinear effect is more
pronounced at shorter wavelength, and therefore might offset the limitations of loss and
dispersion as the pump move to shorter wavelengths.

Table 2. Phase matching diameters and effective indices for THG between fundamental
pump mode and different third harmonic modes for a silica microfiber in air. Pump
wavelength is $\lambda_p = 0.8 \mu m$

<table>
<thead>
<tr>
<th>Fundamental</th>
<th>Harmonic</th>
<th>$D(\mu m)$</th>
<th>$n_{eff}$</th>
</tr>
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<tbody>
<tr>
<td>HE11</td>
<td>EH11</td>
<td>0.3</td>
<td>1.02</td>
</tr>
<tr>
<td>HE11</td>
<td>HE12</td>
<td>0.36</td>
<td>1.06</td>
</tr>
<tr>
<td>HE11</td>
<td>EH12</td>
<td>1.13</td>
<td>1.38</td>
</tr>
<tr>
<td>HE11</td>
<td>HE13</td>
<td>1.24</td>
<td>1.39</td>
</tr>
<tr>
<td>HE11</td>
<td>EH13</td>
<td>2.10</td>
<td>1.43</td>
</tr>
<tr>
<td>HE11</td>
<td>HE14</td>
<td>2.17</td>
<td>1.43</td>
</tr>
<tr>
<td>HE11</td>
<td>HE31</td>
<td>0.35</td>
<td>1.05</td>
</tr>
<tr>
<td>HE11</td>
<td>EH31</td>
<td>0.85</td>
<td>1.33</td>
</tr>
<tr>
<td>HE11</td>
<td>HE32</td>
<td>0.35</td>
<td>1.05</td>
</tr>
</tbody>
</table>

The large variation in maximum efficiency for the different modes can be
understood by investigating the intensity distribution of the different modes, as the
efficiency is highly dependent on the spatial overlap of the electric field distributions of
the two interacting modes. The intensity distribution shown in Figure 5 provides a visual
understanding of the modes spatial overlap. There is a visible significant overlap between
the $HE_{11}$ and $HE_{12}$ modes, while a relatively small overlap between the $HE_{11}$ and $EH_{11}$
modes, hence the observation that the $HE_{12}$ mode being much more efficient than the
$EH_{11}$ mode. This observation can be extended to all the other higher harmonic modes.
Figure 3. The values of the $J_3$ overlap between the fundamental mode at the pump wavelength with for different higher order modes at the harmonic wavelength. Two different pump wavelengths $\lambda_1 = 0.8 \, \mu m$ and $\lambda_1 = 1.064 \, \mu m$ are shown as comparison.
Figure 4. Variation of the values of the $J_3$ overlap and phase matching diameters with wavelength. $J_3$ determines the efficiency of the THG process, and the increase in $J_3$ can be understood to be due to increasing modal confinement as the wavelength is reduced.

The effect of loss on efficiency, included in equations 1 and 2, is shown in Figure 6. The process efficiency can be quite high, up to $\eta = 80\%$, over several mm when coupled with the $HE_{12}(3\omega)$ mode. Even for higher losses of around 0.8 dB/mm, it is still possible to attain 40% conversion, but the harmonic signal falls with distance quickly and hence should be coupled out after 5 mm. This is expected as more of the generated light is absorbed as it traverses longer distances, causing absorption effects to dominate at longer distances. As the regime of loss in which silica operates is very small, the optimal length for THG - shown as the dotted line in Figure 6 - is quite high, although it can be seen that the THG efficiency exceed 60% for distances from 5 mm and above. Shorter
pumping wavelengths allow for higher efficiencies since the tighter maximum modal confinement increases the overlap.

Figure 5. Intensity field distribution for (a) HE_{11} mode for the pump wavelength (0.8 \mu m) and (b) HE_{12} and (c) EH_{11} modes for the harmonic wavelength (0.267 \mu m), both at their respective critical diameter.

However, for real microfibers exhibit other nonlinear effects, i.e. Self-Phase Modulation (SPM) and Cross Phase Modulation (XPM). The change in diameter required to remove these effects are minute and extremely sensitive to variation, making it practically impossible to achieve. The efficiency of more realistic microfibers is represented in figure 7, where oscillations in efficiency are clearly seen. These oscillations are attributed to SPM and XPM effects, encapsulated in \( J_1, J_2 \) and \( J_5 \) in
equations 1 and 2. The reduction in the strength of the oscillations at longer lengths and
at higher losses is due to the depletion of the pump wavelength, which therefore reduces
both SPM and XPM effects, in addition to the harmonic generation.

Figure 6. Effect of UV absorption loss $\alpha_3$ on the efficiency $\eta$ of THG using a $\lambda_1 =$
$0.8 \mu m$ pump, when coupled to the HE$_{12}(3\omega)$ mode with $\alpha_1 = 0$ and $P_1 = 1$ kW for an
ideal microfiber corrected for SPM and XPM effects.
Figure 7. Changes in efficiency $\eta$ of THG due to UV loss, $\alpha_3$ for a pump wavelength of $\lambda_1 = 0.8 \mu m$ coupled to the HE$_{12}(3\omega)$ mode for a realistic microfiber at the phase matching diameter. Calculations were made for a fiber with no loss at the pump wavelength ($\alpha_1 = 0$) and $P_1 = 1 \text{ kW}$.

3.1.2 Experimental result

To demonstrate the feasibility of UV generation via THG, a Ytterbium Doped Fiber Laser MOPA system, shown in figure 8a, with a spectral peak at $1.068 \mu m$, pulse width of $2 \text{ ns}$, repetition rate of $500 \text{ kHz}$, maximum peak power of $5 \text{ kW}$ and maximum average power of $5 \text{ W}$ was employed to investigate the generation of UV at $0.356 \mu m$.

To achieve THG between the $HE_{11}(\omega)$ and $HE_{12}(3\omega)$ modes, a fiber diameter of $0.513 \mu m$ is required to satisfy the phase matching condition. However microfibers with waists slightly smaller than the optical diameter can satisfy the phase matching condition at several locations in the transition region of the microfiber [5]. We therefore fabricated a microfiber with a waist of $\sim 501 \text{ nm}$, which is consistently smaller than the phase
matching diameter. The output spectra are then monitored using an OceanOptics USB4000 spectrometer, and the output power is increased until the taper broke. The resulting spectra are given in Figure 8b.
Figure 8. (a) Schematic of the Ytterbium Doped Fiber (YDF) Master Oscillator Parametric Amplifier (MOPA) used in the THG UV experiment. Note: EOM is Electro-Optic Modulator, AOM is Acousto-Optic Modulator, A1 – A4 are amplifiers. (b) Spectral output for THG for a pump wavelength of $\lambda_1 = 1.068 \mu m$ with 5 different peak powers. (c) Dependence of the peak intensity on input peak power. The exponential fit equation is included.

THG for five different peak powers have been investigated, as the microfiber broke for peak powers exceeding 2.2 kW. As can be seen, the peak is at 356.14 nm. The FWHM is less than 0.5 nm, which is also within experimental error. Figure 8c shows the dependence of the peak power at 356.14 nm on the input power. The peak intensity has a cubic dependence on the input power, which is expected from a $\chi^{(3)}$ process. The inability
to achieve the highest possible efficiency has been attributed to the imperfections in the
diameter uniformity. Regardless of this, THG in microfibers can be a very promising
candidate for efficient generation of light in the UV if optimization can take into account
SPM and XPM effects.

3.2 Mid-IR Generation

In addition to the THG process, $\chi^{(3)}$ nonlinearity can also support parametric
downconversion, known as one third harmonic generation or TPG. This process allows
the amplification of optical waves at longer wavelength by using a pump source at shorter
wavelength via parametric downconversion in the optical microfiber. The TPG process
has been previously analyzed by using highly Germania-doped fibers [18]. However,
because of the choice of materials, the optimized modes at the shorter wavelength can
only be HE$_{13}$, with HE$_{11}$ being the mode at the longer wavelength. The disadvantage of
this is that while the Germanium-doped fibers can potentially be much longer than optical
microfibers, the modal overlap between HE$_{13}$ and HE$_{11}$ is much smaller than that between
HE$_{11}$ and HE$_{12}$, requiring much larger lengths for the same theoretical efficiency.

Here we consider the interaction between $\lambda_3=1.064$ μm (pump) and $\lambda_1=3.192$ μm
(seed) in an optical microfiber. The required phase matching conditions for TPG using a
higher order mode at $3\omega$ to produce one-third ($\omega$) harmonic in the mid-IR at the
fundamental mode is illustrated in Figure 9. As with the case of UV generation, the most
suitable mode for TPG is still the HE$_{12}$. For this mode, the required phase matching
diameter is about 1.55 μm and the $J_3$ overlap integral is about 0.07 μm$^2$. 
Figure 9. Dependence of the effective indices of the pump and one-third harmonic modes against diameter for TPG in a silica microfiber in air. Pump wavelength is $\lambda_1 = 1.064 \mu m$.

However, as mentioned earlier, the TPG process must be seeded to achieve high efficiency. Assuming a pump power of 1 kW at $\lambda_3$ and a seed power of 50 W at $\lambda_1$, the TPG conversion efficiency with different transmission loss at 3.192 $\mu m$ are shown in Figure 10. It was found that the TPG conversion efficiency is more sensitive to transmission losses as compared to THG. Therefore, in order to obtain conversion efficiencies higher than 2%, transmission losses of less than 4 dB/m is preferred. The dependence of TPG performance on seed power is shown in Figure 11. Here, the pump power and loss were fixed at 1000 W and 0.8 dB/m, respectively, while the conversion evolution along fiber length was evaluated for seed powers of 10 W, 20 W and 100 W. The conversion efficiency is clearly highly dependent on the power level of the seed, and high conversion efficiencies are more easily achieved with high seed powers.
Figure 10. Effect of Mid-IR transmission loss $\alpha_1$ on the efficiency $\eta$ of TPG using a $\lambda_3 = 1.064 \mu m$ pump for an ideal microfiber corrected for SPM and XPM effects.
Conclusion

In conclusion, it is possible to exploit $\chi^{(3)}$ processes in silica to generate wavelengths within the UV and mid IR. The generation of both UV and mid IR radiation in optical microfibers requires the phase matching between the fundamental mode of the longer wavelength and a harmonic mode of the shorter wavelength. It was discovered that the most efficient harmonic mode to be the $HE_{12}$ mode. However, other nonlinear effects such as SPM and XPM may be exhibited, requiring a slight detuning from the original phase matching diameter. The efficiency of such processes have been shown to be theoretically very high, and several experimental work have demonstrated the possibility of using this mechanism as a method of obtaining coherent radiation at wavelengths otherwise accessible via conventional means.
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