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Efficient phase-matched third harmonic generation in an asymmetric plasmonic slot waveguide

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Abstract: An asymmetric plasmonic slot waveguide (APSW) for efficient phase-matched third harmonic generation (THG) is proposed and demonstrated theoretically. Nonlinear organic material DDMEBT polymer is integrated into the bottom of the metallic slot, while silicon is used to fill the top of the slot. We introduce the rigorous coupled-mode equations of THG in the lossy APSW and apply them to optimize the waveguide geometry. Taking advantage of the surface plasmon polaritons (SPPs), the electric fields can be tightly confined in the metallic slot region and the nonlinear effect is greatly enhanced accordingly. Then, we investigate the relationships between THG efficiency and parameters such as slot width and height, phase matching condition (PMC), modal overlap related nonlinear parameter, figure-of-merit, pump power and detuning. With the proposed asymmetric waveguide, we demonstrate a high THG conversion efficiency of $4.88 \times 10^{-6}$ with a pump power of 1 W and a detuning constant of $-36 \text{ m}^{-1}$ at a waveguide length of 10.65 $\mu$m.

OCIS codes: (070.4340) Nonlinear optical signal processing; (190.2620) Harmonic generation and mixing; (250.5403) Plasmonics; (250.4390) Nonlinear optics, integrated optics.

References and links


1. Introduction

During the past few years, mid-infrared (mid-IR) photonics have been attracting increasing attentions [1, 2]. Accompany with the development of this special waveband, various all-optical integrated functional devices in the mid-IR region, such as amplifiers and wavelength converters, have been studied for potential applications in free space communications, chemical and biological sensing, and medical procedures [3, 4]. However, there is still little investigation on passive photonic devices based in mid-IR waveband to realize the highly desired signal processing functions such as dispersion monitoring, switching and wavelength conversion. In 1.55 μm region, third harmonic generation (THG) has been demonstrated to be a promising way to realize high speed optical performance monitoring of...
optical signal to noise ratio (OSNR) and residual dispersion [5]. Due to the development of mid-IR communication [6, 7], THG devices working in this special waveband can find applications in the area of signal processing. Therefore, design for efficient THG devices working in the mid-IR waveband is interesting and valuable.

The basic theory of the THG was developed by Armstrong et al. [8] and extended in [9], in which the analytical solutions and conversion efficiency of THG were investigated. In a Kerr nonlinear material, such as silica nitride (Si$_3$N$_4$), it is possible to generate third harmonics with an incident wave. The key to improve the nonlinear conversion efficiency depends on the nonlinearity of the interactive material, modal overlap, and phase-matching conditions (PMC), which can be satisfied by employing advanced devices or new types of nonlinear materials. Recently, the development of ultra-high density circuitry based on surface plasmon polaritons (SPPs) provides an effective solution [10]. Thanks to the characteristics of SPPs, the light can be tightly confined in the slot region beyond the limit imposed by the laws of diffraction in dielectric medium [11, 12]. More recently, several types of plasmonic waveguide structures have been proposed, such as metallic grooves [13] and metal-insulator-metal structures [14]. However, the metal-induced linear propagation loss is still a concern in these plasmonic waveguides. To realize the trade-off between propagation loss and subwavelength mode confinement, slot waveguide concept has been proposed [15]. Generally, one of the advantages of THG in plasmonic slot waveguides is that the nonlinear properties can be engineered through appropriate design of the waveguide geometric parameters. Furthermore, PMC, being another key factor, can be satisfied between the pump field and the generated third harmonic field by tailoring the mode dispersion in these waveguides. In addition, the dielectric medium in the metallic slot region can be organic nonlinear optical materials that exhibit high third-order nonlinear susceptibilities. Recently, Beels et al. have proposed a new type of material with high optical quality for third-order integrated nonlinear optics - DDEMBT polymer [16]. Furthermore, DDEMBT polymer is free from two-photon absorption (TPA). Hence, slot waveguide structures combining the plasmonic and DDEMBT polymer have huge potential for nonlinear nano-optics.

What we need to be reminded here is the necessity of the asymmetric slot waveguide structure to the highly efficient THG. In general, a slot waveguide supports several modes, and the effective index of the fundamental mode is larger than the higher-order mode at a specific frequency. Therefore, PMC can be fulfilled if the fundamental frequency (FF) propagates at a lower-order mode, known as the inter-modal phase-matching technique [17, 18]. In general, the FF always assumed to propagate at fundamental waveguide mode. Furthermore, to generate third harmonic wave, the modal overlap between the fundamental mode at FF and each higher-order mode at third harmonic frequency (THF) must be greater than zero. Therefore, THF should propagate at the 2-nd, 4-th or higher even order modes due to the symmetry of the common planar slot waveguides. For example, zero modal overlap happens between the fundamental mode at FF and the 1-st mode at THF in a common planar slot waveguide, because the distribution of the fundamental waveguide mode at FF is nearly symmetrical, while the distribution of the 1-st waveguide mode at THF is nearly anti-symmetrical [19]. Due to the higher even order mode at THF, inter-modal PMC results in very small modal overlap between the two considered modes despite its advantage of flexibility. In order to circumvent this problem and enlarge the modal overlap, we introduce an asymmetric plasmonic slot waveguide structure by employing different cladding and substrate materials.

In this paper, we propose for the first time a new kind of third-order nonlinear asymmetrical plasmonic slot waveguide (APSW) configuration to break the symmetry of the higher-order mode at THF and choose DDEMBT polymer with high $\chi^{(3)}$ and silicon as its core dielectric materials. This waveguide is capable of tightly confining electric fields in the nonlinear slot region both at FF and THF, which can efficiently enhance the nonlinear effects. The PMC between the fundamental pump mode and the higher-order third harmonic mode in the APSW can be satisfied with the inter-modal phase-matching technique. Finally, the relationships between...
THG efficiency and parameters such as slot width and height, PMC, modal overlap related nonlinear parameter, figure-of-merit, pump power and detuning are studied in details to provide a guideline to achieve efficient THG.

2. Basic THG theory in the APSW

Figure 1 shows the cross-section view of the proposed APSW. The width and height of the metallic slot are $w$, $h_o$ (DDMEBT polymer slot height) and $h_s$ (silicon slot height), respectively. Silver (Ag) is considered as the metal cladding medium to form the SPPs due to its relatively low induced linear propagation loss in the calculated wavelength range and its thickness $h_{Ag}$ is fixed to be 500 nm. The substrate is formed by silica ($\text{SiO}_2$). This asymmetric waveguide structure is able to greatly increase the modal overlap between the guided waveguide modes during the third-order nonlinear process. Furthermore, such structure can be realized using fabrication techniques similar to those of conventional plasmonic slot waveguides. First, a thin silicon layer is deposited on a standard silicon-on-insulator wafer using plasma enhanced chemical vapor deposition and then patterned utilizing electro-beam lithography followed by reactive ion etching. The DDMEBT can be done by ion implantation. Finally, a thin silver layer is deposited on top by sputtering. To couple light into a metal-clad plasmonic slot waveguide from a standard silicon waveguide, X. Sun et al. proposed adiabatic tapers at the two plasmonic waveguide ends, which achieved a 91% coupling efficiency [20]. The adiabatic tapers are also covered with silver to enable a smooth mode transformation.

It should be noted that, the nonlinear response of silver in the waveguide is neglected during the THG process, since the electric field does not penetrate deeply inside the metal and decays exponentially with distance from the metal-dielectric interface. Therefore, to study the THG process in the considered APSW, the nonlinear coupled mode theory for waveguides with substantial loss is outlined. To correctly model the nonlinear optical interactions, all components of the excited electric and magnetic fields in the APSW should be taken into consideration. From Maxwell’s curl equations,

$$\nabla \times \vec{E}(r,t) = -\mu \frac{\partial \vec{H}(r,t)}{\partial t}$$  \hspace{1cm} (1)
\[ \nabla \times \mathbf{H}(r,t) = \varepsilon \frac{\partial \mathbf{E}(r,t)}{\partial t} + \frac{\partial \mathbf{P}_{NL}}{\partial t} \]

(2)

where \( \varepsilon \) and \( \mu \) are the linear permittivity and permeability. \( \mathbf{P}_{NL} \) is the nonlinear polarization vector and \( \frac{\partial \mathbf{P}_{NL}}{\partial t} \) can be viewed as a source term that arises from the nonlinear interaction. Assuming only two different propagating modes in the APSW, i.e., pump at frequency \( \omega_0 \) and its third harmonics at \( 3\omega_0 \), neglecting their linear losses at first, the total excited electric fields \( \mathbf{E} \) and magnetic fields \( \mathbf{H} \) at any location of the waveguide can be described as below [21, 22]:

\[ \mathbf{E}(r,t) = \frac{1}{2} \sum_j A_j(z) Z_j^{1/2} F_j(r_{\perp}) \exp[i(\beta_j z - \omega_j t)] + c.c. \]

(3)

\[ \mathbf{H}(r,t) = \frac{1}{2} \sum_j A_j(z) Z_j^{1/2} G_j(r_{\perp}) \exp[i(\beta_j z - \omega_j t)] + c.c. \]

(4)

where \( \beta_j = \frac{\omega_j}{c} n_{eff}(\omega_j) \) is the propagation constant, \( A_j(z) \) is the slowly-varying mode amplitude, \( F_j(r_{\perp}) \) and \( G_j(r_{\perp}) \) are mode profiles which have been normalized with

\[ \frac{1}{4} \int_{A_{NL}} (\mathbf{F}_j \times \mathbf{G}_j + \mathbf{F}_j^* \times \mathbf{G}_j^*) \cdot dxdy = 1, \]

\( j = 1 \) refers to the case of FF, and \( j = 3 \) refers to the case of THF. \( Z_0 = \sqrt{\mu_0/\varepsilon_0} \) here is used to simplify the numerical calculation. \( r = (x, y, z) \) and \( r_{\perp} = (x, y) \). With the normalization, the corresponding field power can be expressed as \( P_j(z) = |A_j(z)|^2 \).

We apply the reciprocity theorem to calculate the derivatives of the amplitudes \( dA_j/dz \) and obtain the following:

\[ \frac{dA_j}{dz} = \frac{Z_0^{1/2}}{2} \int_{A_{NL}} \left\{ \exp[-i(\beta_j z - \omega_j t)] F_j^* \frac{\partial \mathbf{P}_{NL}}{\partial t} \right\} dxdy \]

(5)

where \( \langle \cdot \cdot \rangle \) is time averaging, \( A_{NL} \) is the cross-section of the waveguide, and the nonlinear polarization related to the electric field \( \mathbf{E} \) is described as [19]:

\[ \mathbf{P}_{NL} = \varepsilon_0 \chi^{(3)}(r) (\mathbf{E}(r,t) \cdot \mathbf{E}(r,t)) \mathbf{E}(r,t) \]

(6)

For lossy waveguides case, the complex propagation constant can be written as: \( \beta = \beta_j + i \alpha_j/2 \), where \( \beta_j \) and \( \alpha_j \) are both real and positive. They represent the phase propagation constant and linear propagation loss coefficient, respectively. We define

\[ A_j = \tilde{A}_j \exp(-\frac{\alpha_j z}{2}) \]

(7)

and substitute it into Eq. (6), the nonlinear polarization can be rewritten as:

\[ \mathbf{P}_{NL} = \varepsilon_0 \chi^{(3)}(r) (\tilde{\mathbf{E}}(r,t) \cdot \tilde{\mathbf{E}}(r,t)) \tilde{\mathbf{E}}(r,t) \]

(8)
where \( E(\mathbf{r},t) = \frac{1}{2} \sum_j A_j(z) Z_j^{ij} \bar{F}_j(\mathbf{r}_\perp) \exp[i(\mathbf{P}_j z - \omega_j t)] + \text{c.c.} \). We can then obtain the following nonlinear coupled-wave equation for the lossy APSW:

\[
\frac{dA}{dz} = -\frac{Z_0^{ij}}{2} \exp(\frac{\alpha z}{2}) \int \left\{ \exp[-i(\mathbf{P}_j z - \omega_j t)] \bar{F}_j \cdot \frac{\partial P_{\omega_2}}{\partial t} \right\} d\mathbf{r}_i d\mathbf{r}_2
\]  

(9)

Note that the THG nonlinear process in silicon crystals is based on the third-order susceptibility \( \chi^{(3)} \). However, the third-order susceptibility of silicon also leads to TPA when the energy of photos exceeds the half band gap \( E_g/2 \), where \( E_g/2 \) corresponds to a wavelength of 2.2 \( \mu m \). The real and imaginary parts of \( \chi^{(3)} \) are related to the nonlinear refractive index \( n_2 \) and the TPA coefficient \( \alpha_2 \) by

\[
\frac{\omega_3}{c} n_2 + \i \frac{\alpha_2}{2} = \frac{3\omega}{4\epsilon_0 \epsilon_0^* n_0^2(\omega)} \chi^{(3)} \quad [23],
\]

where \( n_0(\omega) \) is the linear refractive index of silicon at different frequencies. Since the TPA coefficient decreases with the increase of wavelengths longer than 1.7 \( \mu m \) and eventually drops to 0 for wavelengths longer than 2.2 \( \mu m \) for silicon, we focus the TPA effect in silicon on the generated third harmonic wave.

Under the continuous wave (CW) condition, by taking the TPA effect into consideration, we can obtain the two nonlinear equations describing the THG process in the APSW:

\[
\frac{\partial A}{\partial z} = i (|I_1| A_1 + |I_2| A_2) A_1 + i (|I_3| A_3^* A_1 e^{\i \delta \beta}) \exp(\frac{\alpha z}{2})
\]  

(10)

\[
\frac{\partial A}{\partial z} = i (3|I_4| A_4^* A_1 + |I_5| A_5) A_1 + i (|I_6| A_6^* e^{\i \delta \beta}) \exp(\frac{\alpha z}{2})
\]  

(11)

Here \( \delta \beta = \beta_3 - 3\beta_1 \) is the phase-mismatch, \( \alpha_1 \) is the linear propagation loss coefficient and the nonlinear parameter related overlap integrals \( I_1, I_2, I_3, I_4, I_5 \) and \( I_6 \) are defined as:

\[
I_1 = \frac{1}{12} \oint_{\mathbf{A}_{st}} (2 |\bar{F}_1|^2 + |\bar{F}_2|^2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(12)

\[
I_2 = \frac{1}{6} \oint_{\mathbf{A}_{st}} (|\bar{F}_1|^2 |\bar{F}_2|^2 + |\bar{F}_1 \cdot \bar{F}_2|^2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(13)

\[
I_3 = \frac{1}{4} \oint_{\mathbf{A}_{st}} (|\bar{F}_1|^2 (\bar{F}_1 \cdot \bar{F}_2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(14)

\[
I_4 = \frac{1}{6} \oint_{\mathbf{A}_{st}} (|\bar{F}_1|^2 |\bar{F}_1|^2 + |\bar{F}_1 \cdot \bar{F}_2|^2 + |\bar{F}_1 \cdot \bar{F}_3|^2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(15)

\[
I_5 = \frac{1}{4} \oint_{\mathbf{A}_{st}} (|\bar{F}_1|^2 + |\bar{F}_2|^2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(16)

\[
I_6 = \frac{1}{4} \oint_{\mathbf{A}_{st}} (|\bar{F}_1|^2 |\bar{F}_2|^2) \cdot n_2^2(\omega, \mathbf{r}_\perp) \cdot [k, n_2(\mathbf{r}_\perp)] dS
\]  

(17)
where $n_z(\vec{r}_z)$ and $\alpha_z(\vec{r}_z)$ are the nonlinear refractive index and TPA coefficient at any location of the waveguide, $k = 2 \pi / \lambda$ is the wave number, $n_0(\omega, \vec{r}_z)$ is the linear refractive index at FF and THF in any position of the waveguide. Here $\alpha_z = 5.3 \times 10^{-12}$ m/W is the TPA coefficient for silicon at the wavelength of 1200 nm [24]. For DDMEBT polymer, $n_2$ (DDMEBT) = $8.5 \times 10^{-18}$ m²/W [16], while for silicon, $n_2$ (Si) = $4.5 \times 10^{-18}$ m²/W [25]. We assume the nonlinear refractive index $n_z(\vec{r}_z)$ to be constant in a specific medium. By substituting Eq. (7) into Eqs. (10) and (11), we finally obtain the coupled-mode equations describing the THG process in lossy APSW as:

$$\frac{\partial A_1}{\partial z} = -\frac{\alpha_z}{2} A_1 + i[(I_1 |A_1|^2 + I_2 |A_2|^2) A_1 + I_3(A_3^* A_1 e^{i\delta \beta})]$$

(18)

$$\frac{\partial A_4}{\partial z} = -\frac{\alpha_z}{2} A_4 + i[(3I_3 |A_3|^2 + I_1 |A_1|^2) A_4 + I_1(A_1^* A_3 e^{-i\delta \beta})]$$

(19)

Using the substitution of $A_1 = \sqrt{P_1} e^{i\phi_1}$ and $A_3 = \sqrt{P_3} e^{i\phi_3}$, and defining the pump-harmonic phase difference during the THG process as $\Psi = \delta \beta z + \phi_3 - 3\phi_1$, Eqs. (18) and (19) can be rewritten as:

$$\frac{dP_1}{dz} = -\alpha_z P_1 - 2I_1 P_1^{\frac{3}{2}} P_3^{\frac{1}{2}} \sin \Psi$$

(20)

$$\frac{dP_3}{dz} = -\alpha_z P_3 + 2I_3 P_3^{\frac{3}{2}} P_1^{\frac{1}{2}} \sin \Psi$$

(21)

$$\frac{d\Psi}{dz} = \delta \beta + 3(I_3 - I_1) P_3 + (I_1 - 3I_2) P_1 + (I_6 P_1^{\frac{1}{2}} P_3^{\frac{1}{2}} - 3I_1 P_3^{\frac{1}{2}} P_1^{\frac{1}{2}}) \cos \Psi$$

(22)

It is found that $P_3$ can be neglected in Eq. (22) because of its low value as compared to pump power, i.e. $P_1$. Then Eq. (22) can be rewritten as $\frac{d\Psi}{dz} = \delta \beta + 3(I_3 - I_1) P_1$. This new equation depicts that $\Psi$ is determined mainly by detuning term and pump power term. If the right hand side of Eq. (22) is kept approximately to be 0, power can be transferred from pump ($P_1$) to third harmonic ($P_3$) efficiently. We will use the Runge-Kutta method in MATLAB to numerically calculate Eqs. (18) and (19) in the section 4 to investigate the THG efficiency.

In the end, the definition of the conversion efficiency of THG is given as $\eta = P_3(L_p)/P_1(0)$, where $P_1(0)$ stands for the pump power at FF, $L_p$ stands for the waveguide length when THF reaches its maximum output power $P_3(L_p)$, respectively. Note that, the value of $\delta \beta = \beta_3 - 3\beta_1$ can directly influence the conversion efficiency. Phase-mismatch ($\delta \beta \neq 0$) always results in cycle flowing of the energy between FF and THF and limits the one-way conversion efficiency which makes the THG impossible. Recently, the quasi-phase matching (QPM) technique was theoretically proposed to realize PMC by adopting index grating [26], but it leads to large mode sizes and complexity in structure fabrications. Fortunately, the APSW supports several modes and the effective index of the fundamental mode is larger than the higher-order mode at any specific frequency. PMC then can be appropriately obtained if the FF propagates at a lower-order mode. In our proposed configuration, inter-modal phase-matching technique as described above is adopted [17, 18].
3. Key factors to increase the THG efficiency

In fact, what limits the benefit of using the inter-modal phase-matching technique is that, the spatial modal overlap between guided modes with different orders is always less than that between modes with same orders. This results in a very small modal overlap due to the definition of the modal overlap related nonlinear parameter $\text{Re}(\Gamma_6)$ in Eq. (17), which directly relates to the power transfer strength from FF to THF. In particular, $\text{Re}(\Gamma_6)$ in a common symmetric planar waveguide will be almost 0 if the FF propagates at 0-th mode and the THF propagates at 1-st mode at the same time. In order to enlarge the modal overlap between the considered guided modes, we introduce asymmetric plasmonic slot waveguide by using metal cladding as shown in Fig. 1. This new waveguide structure is compatible with silicon photonics. The number of the guided modes and the corresponding field profiles are determined by its specific waveguide geometry and the wavelength at FF. The other key factors to increase the THG efficiency are reducing the linear propagation loss, satisfying the PMC, and optimizing the figure-of-merit (FOM). In order to reduce linear propagation loss in our work, silver (Ag) was chosen as the metal medium. It is characterized by a Drude permittivity dispersion of $\varepsilon_{Ag} = \varepsilon_\infty - \frac{f_p}{f_0} \left[ \frac{1}{(f + i\gamma)^2} \right]$, with $\varepsilon_\infty = 5$, $f_p = 2175$ THz, and $\gamma = 4.35$ THz [27]. The considered Kerr-type DDBMBT polymer with refractive indices of $n_0 (3600 \text{ nm}) = 1.7277$ and $n_0 (1200 \text{ nm}) = 1.8065$ [16] is integrated into the bottom of the slot region. While a relative thin layer of silicon (Si) at the top of the slot is used to isolate the field from the metal on the top to the DDBMBT polymer, thus making the PMC achievable. The refractive indices of silicon at the working wavelength are $n_{Si} (3600 \text{ nm}) = 3.4274$ and $n_{Si} (1200 \text{ nm}) = 3.5202$.

We first fixed the slot width to be $w = 40$ nm and silicon slot height to be $h_s = 100$ nm, respectively. The effective indices of the 0-th mode at FF ($\lambda_1 = 3600$ nm) and 2-nd mode at THF ($\lambda_1 = 1200$) are calculated by using finite-element-based commercial COMSOL software, with an element size of 4 nm for slot and 80 nm for other region. Figure 2(a) shows the effective indices as a function of DDBMBT polymer slot height $h_o$. We found that, with a wide range of the DDBMBT slot height from 460 nm to 540 nm, the indices of the two guided modes do not change too much. For simplicity, we consider a DDBMBT polymer slot height of 500 nm and study the possibility of achieving PMC by adjusting the height of the silicon slot. Figure 2(b) plots the effective indices of the two guided modes as a function of the silicon slot height for $w = 40$ nm and $h_o = 500$ nm. We can see that the PMC occurs when the silicon slot height is set to be $h_s = 207$ nm. At this point, the effective indices of FF and THF are 2.5663 + 0.0325i and 2.5661 + 0.0089i, respectively. Here, for FF wave, the supported 0-th mode results from the coupling between the fundamental mode in the DDBMBT polymer slot and fundamental mode in the silicon slot. On the other hand, for THF wave, the 2-nd mode results from the coupling between the fundamental mode in the DDBMBT polymer slot and 1-st mode in the silicon slot.

The corresponding dominant mode distributions for the 0-th mode at FF and 2-nd mode at THF are plotted in Figs. 3(a) and 3(b), respectively. The arrows in the 2D field panels reveal the quasi-TE like nature of the two guided modes. It can be found that the mode profile for the 2-nd mode at THF is very asymmetrical. To obtain a more comprehensive understanding of the characteristics about the mode profiles, we plot the $E_x$ distributions along x cutline at $y = 0$ [as the longitudinal dash lines shown in Figs. 3(a) and 3(b)] and y cutline at $x = 0$ [as the horizontal dot lines shown in Figs. 3(a) and 3(b)] in Figs. 3(c) and 3(d), respectively. Both of the modes of interest here are tightly confined in the metallic slot region as shown in Fig. 3(c), which also proves the THG in silver is negligible. Figure 3(d) depicts that, due to the special physical structure of the waveguide, the modal overlap between the two guided modes is very large. Here, the linear propagation losses of FF and THF are 0.493 dB/μm and 0.407 dB/μm, respectively.
As PMC is demonstrated, we next discuss the optimal waveguide geometry for THG since the specified $w$, $h_{s}$, and $h_{p}$ mentioned above are not the only geometrical case satisfying PMC. Figure 4(a) gives the distributions of the DDMEBT polymer slot height and silicon slot height as a function of the slot width satisfying the PMC. As the slot width varying from 40 nm to 60 nm, it is possible to obtain PMC by just optimizing the silicon slot height with a fixed DDMEBT polymer slot height of 500 nm. When the slot is wider than 60 nm, it is impossible to reach the PMC between the 0-th mode at FF and 2-nd mode at THF by just optimizing the silicon slot height. Take slot width of $w = 70$ nm as an example. At first, we set a certain...
DDMEBT slot height to be 500 nm. By regulating the silicon slot height, we can get a minimum difference between the two guided mode indices when \( h_s = 233 \) nm. Note that, the 2-nd mode pattern will not exist if \( h_s \) continues to increase. Therefore, by changing the DDMEBT polymer slot height to be \( h_o = 560 \) nm, PMC can be satisfied as shown in Fig. 4(a).

To assess which of the phase-matched points depicted in Fig. 4(a) would be the most efficient one in THG process, modal overlap related \( \text{Re}(I_6) \) is plotted as a function of the slot width in Fig. 4(b). \( \text{Re}(I_6) \) decreases with increasing slot width due to the weaker field confinement with larger slot area. However, in the proposed lossy APSW, besides \( \text{Re}(I_6) \), TPA in silicon slot and linear propagation loss are the other two aspects which must be taken into consideration. According to our calculation, the effect of TPA in silicon is much less than the linear propagation loss within the slot. We then ignore TPA in this part and define the figure-of-merit (FOM) for phase-matched THG process as 
\[
\text{FOM}_\text{FF, THF} = \frac{\text{Re}(I_6)}{\alpha_j}, j = 1, 3,
\]
and \( \alpha_j \) refers to the linear propagation loss coefficients at FF and THF, respectively. The FOMs at different PMCs are illustrated in Fig. 4(c). Both FOMs decrease with increasing slot width, which indicates that the APSW has better THG performance with narrower slot. Accordingly, the most promising waveguide geometry is \( w = 40 \) nm, \( h_o = 500 \) nm, \( h_s = 207 \) nm, with \( \text{Re}(I_6) = 611.39 \text{ m}^{-1} \cdot \text{W}^{-1} \), \( \text{FOM}_\text{FF} = 0.00124 \text{ W dB}^{-1} \), and \( \text{FOM}_\text{THF} = 0.0015 \text{ W dB}^{-1} \), respectively.

![Fig. 4.](image)

**Fig. 4.** (a) DDMEBT polymer slot height and silicon slot height, (b) overlap integral related \( \text{Re}(I_6) \), and (c) FOM of FF and THF versus the metal plasmonic slot width at different PMCs

### 4. Simulation results

Now we numerically study the nonlinear THG performance in the proposed APSW. Assuming a pump power of 1 W, the conversion efficiency and the corresponding waveguide length \( L_p \) at different PMCs are illustrated in Fig. 5. The conversion efficiency \( \eta \) increases together with shorter waveguide length with the reduction of the slot width. These results are consistent with the FOM variation as discussed above. Note that the pump power can be obtained in practice by
typical mid-IR lasers, such as CW optical parametric oscillators [28–34]. In Fig. 6(a), we give the efficiency contour map with different detuning constants for the waveguide geometry as shown in Fig. 3. A maximum conversion efficiency of $4.88 \times 10^{-6}$ happens with an optimized detuning of $\delta \beta = -36 \text{ m}^{-1}$. The value is negatively offset from zero to compensate for the nonlinear phase shift during the nonlinear process, although a smaller but nonetheless significant third harmonic signal would still be achieved if $\delta \beta = 0 \text{ m}^{-1}$. Figure 6(b) shows the evolution simulation results of the FF and THF powers along the propagation distance with a fixed pump power of $P_1 = 1 \text{ W}$ and a detuning of $\delta \beta = -36 \text{ m}^{-1}$. One can see that the power of the fundamental mode at FF decreases monotonously due to the nonlinear conversion process and its linear propagation loss. The nonlinear conversion mainly contributed to the THF, which increases first before decreases and reaches its maximum power of $4.88 \times 10^{-6} \text{ W}$ at a 10.65 $\mu\text{m}$ waveguide length. This short waveguide length is also crucial in overcoming other competing nonlinear effects (such as stimulated Raman and Brillouin scattering), which increase exponentially with propagation distance. We also examine the conversion efficiency and the corresponding optimized detuning as a function of pump power, as shown in Fig. 6(c). The optimized detuning decreases from $-6 \text{ m}^{-1}$ to $-36 \text{ m}^{-1}$ as pump power increases from 0.2 to 1 W, which conforms to the Eq. (22). The conversion efficiency increases with increasing pump power and can be further increased with pump power higher than 1 W.

According to our calculation, the value of $\text{Re}(I_3)$, FOM_FF, FOM_THF, and conversion $\eta$ all increase with the reduction the slot width as shown in Figs. 7(a)-7(c). It can be summarized that the THG performance will be further improved with smaller slot width. However, slots that are too narrow should not be considered due to the difficulty in fabrication.

![Graph](image.png)

**Fig. 5.** Conversion efficiency of THG and the corresponding waveguide length Lp as a function of the slot width at different PMCs for a fixed pump power of 1 W.
Fig. 6. (a) Contour map of conversion efficiency in MPSW with different $\delta\beta$ and (b) optical powers of FF and THF along the propagation distance with $\delta\beta = -36$ m$^{-1}$ with a fixed pump power of 1 W; (c) conversion and the corresponding detuning versus the pump power.

Fig. 7. (a) Re($I_6$), (b) FOMs, and (c) conversion efficiency as a function of the slot width.
5. Conclusion

In the present work, we have proposed a new asymmetric plasmonic slot waveguide (APSW) for the first time by considering DDMEBT polymer integrated into the bottom of the metallic slot while silicon filled the top and theoretically investigate the possibility of achieving efficient third harmonic generation (THG). For the THG induced in this APSW, the phase matching condition (PMC) between fundamental frequency (FF) and third harmonic frequency (THF) can be satisfied by appropriately designing the waveguide slot geometrical parameters. Parameters such as slot width and height, phase matching condition (PMC), modal overlap related nonlinear parameter, figure-of-merit, pump power and detuning for THG were investigated in details. In the specially optimized waveguide, with a pump power of 1 W and a detuning constant of $-36 \text{ m}^{-1}$, a very high THG conversion efficiency of $4.88 \times 10^{-6}$ can be obtained at a $10.65 \mu\text{m}$ waveguide length.

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