Second harmonic generation using an electrically controlled asymmetric plasmonic waveguide

Mohamadreza Soltani and Mahmoud Nikoufar

Department of Electrical Engineering, Tiran Branch, Islamic Azad University, Tiran, Iran; Department of Electronics, Faculty of Electrical and Computer Engineering, University of Kashan, Kashan, Iran

ABSTRACT

In this study, it was shown that field enhancement in the nonlinear metal-insulator-metal (MIM) plasmonic waveguides can result in a large enhancement of the second harmonic generation (SHG) magnitude as compared with values reported in the literature. The proposed structure has two metals at the top and two metals at the bottom of the crystal. In this structure, a voltage is applied on metals to produce a SHG electrically. Hence, the metals that define the cavity also serve as electrodes capable of generating high direct current electric fields across the nonlinear material. The frequency of a fundamental wave at 458 nm was doubled and modulated in intensity by applying a moderate external voltage to the electrodes, yielding a voltage-dependent nonlinear generation with a higher coupling efficiency. All the simulations here have been calculated by using the finite-element-based commercial COMSOL software.

1. Introduction

In recent years, plasmonic devices have attracted considerable attention [1–4]. Generally, plasmonics can be considered as light on metal-dielectric interfaces. In this condition, electrons at the surface of metal are collectively accelerated and decelerated by the electric field of light with high frequency. This activity of electrons is similar to plasma. As electrons are involved in the propagation of light, it can be localised into sub-wavelength dimensions [5]. Particularly, a surface wave would be formed by oscillations of collective electrons that exponentially decay into two adjacent half spaces. This collective electronic surface wave that oscillates with the frequency of light is called surface plasmon mode. Although the field decays exponentially into two half spaces, it has an imaginary wave phase, which is propagated along the surface.

These accelerated electrons of metal may increase the momentum of light at the surface. Therefore, the apparent wavelength of light may be reduced, and as a result, field localisation beyond the diffraction limit of light can be possible. Therefore, the optical mode can be confined to the sub-wavelength scale, and the size of the optical mode can be minimised. For example, light can be confined to a size 100 times smaller than its
wavelength [3,4,6–8]. This surface localisation makes plasmonic waveguides an intriguing alternative to conventional dielectric waveguides.

Plasmonic waveguides, based on insulator-metal-insulator waveguides, have been studied extensively as well [9,10]. However, metal-insulator-metal (MIM) waveguides have higher confinement factors and closer spacing to adjacent waveguides [11,12] and have been proposed for a lot of applications such as optical resonators, waveguide bends and splitters based on MIM sub-wavelength plasmonic waveguides [13–15].

The MIM waveguides studied previously have symmetric structure, meaning that the metals at the top and bottom of the dielectric layer are the same [14,15]. Also, asymmetric MIMs have not been investigated, and to the best of our knowledge, a few researchers have investigated the nonlinear optical phenomena in these asymmetric waveguides. Moreover, little attention has so far been paid to the second harmonic generation (SHG) in plasmonic waveguides, and this issue on MIM plasmonic waveguides is yet to be studied extensively. Among the nonlinear processes, SHG is frequently studied because of its interesting applications and simple theoretical principle [16]. In silicon, however, SHG cannot be directly excited, because the second-order susceptibility vanishes in this material, as a result of the crystal centro-symmetry. Exploitation of silicon nitride (Si$_3$N$_4$) has been proposed to solve this drawback and induces second-order nonlinear processes in silicon compatible structures [17–20]. More recently, Oliveira et al. reported SHG in a 20 μm radius Si$_3$N$_4$ ring resonator by using the electric field induced SHG process and calculated a conversion efficiency of about 3.68 × 10$^{-3}$ with a pumping power of 75 mw [19]. In order to further increase the efficiency of SHG and reduce the sizes of devices, the lithium niobate (LiNbO$_3$) crystal in plasmonic-based nonlinear devices are among the most promising candidates to match this expectation, as a result of their ability to allow strong local-enhanced confinement of light beyond the limits imposed by the laws of diffraction in dielectric media [20]. Several kinds of nonlinear plasmonic structures have been proposed. For instance, efficient SHG has been presented in plasmonic slot waveguides (PSW) [21], long-range plasmonic waveguides [22], hybrid plasmonic waveguides (HPW) [23], metal surfaces with nanoscale roughness [24], individual metallic nan-aperture [25], plasmonic particle chains [26] and plasmonic core-shell nanowires [27]. From our knowledge, the most used nonlinear material in these structures to realise SHG is LiNbO$_3$ [16]. However, in spite of the generally used continuous wave pump power of the fundamental frequency (FF) to about 1 W, the peak powers of the generated SHF are usually limited to 10$^{-5}$ W [17]. Even if it can be increased to 10$^{-2}$ W in HPW, the corresponding waveguide length to realise this efficiency is 1 mm [18], which is probably too long and not suitable for application in future integrated nanophotonic circuits. The rather small reported efficiencies are due to the relatively small nonlinear susceptibility in crystal, the moderately large nonlinear coupling coefficients between different frequencies and the absorption loss of the plasmonic modes. In this study, it was shown that field enhancement in asymmetric MIM waveguides can result in large enhancement of SHG magnitude compared to the literature values. The proposed structure has two metals on top and two metals on bottom of crystal. In proposed structure, a voltage is applied on metals to produce SHG electrically. Therefore, the metals that define the cavity also serve as electrodes that can generate high direct current electric fields across the nonlinear material. A fundamental wave at 458 nm
was frequency doubled and modulated in intensity by applying a moderate external voltage to the electrodes, yielding higher coupling efficiency.

2. The structure of MIM waveguide

Figure 1 shows the MIM plasmonic waveguide structure under consideration. As the figure shows, a crystal slot with thickness $a$, which is 50 nm, is sandwiched by top and bottom metals. The top and bottom metals consist of two different metals. In fact, the top and bottom metals of waveguide are departed in two parts. The lengths of two parts are $d_1$ and $d_2$. In this paper, the ratio of $d_1$ and $d_2$ is optimised to reach the highest SHG. One of the following metals gold (Au), silver (Ag) or aluminium (Al) is used as metal$_1$ and metal$_2$. As reported, a symmetric plasmonic waveguide can sustain two different modes [5]. A number of different terminologies such as symmetric and asymmetric modes, long-range surface plasmon polariton (LRSP) and short-range surface plasmon polariton (SRSP), as well as even and odd modes are used in literature to distinguish between the two modes. Concerning asymmetric plasmonic waveguides, two different modes of quasi-symmetric and quasi-asymmetric can exist. Similar to the case of symmetric waveguides, this terminology is based on charge distribution across the centre layer of the waveguide. The profile of LRSP and SRSP modes in asymmetric structures is different from those in symmetric structures. The asymmetric modes can cause the crystal to be centro-asymmetric as a result SHG to be excited in this structure. It has been shown that asymmetric structures have higher SHG enhancement factor. As is shown in the figure, a voltage is applied on metals to produce SHG electrically. Therefore, the metals that define the cavity also serve as electrodes that can generate high direct current electric fields across the nonlinear material. A fundamental wave at 458 nm was frequency doubled and modulated in intensity by applying a moderate external voltage to the electrodes.

In this paper, the wavelength operation of $\lambda = 458$ nm was considered. At this wavelength, the permittivity of crystal (LiNbO$_3$) is $\varepsilon_z = 5.1772$ and $\varepsilon_x = 5.6243$, the
permittivity of aluminium is $\varepsilon_{\text{Al}} = -29 + 7i$, the permittivity of gold is $\varepsilon_{\text{Au}} = -1.3 + 4.6i$ and the permittivity of silver is $\varepsilon_{\text{Ag}} = -6 + 0.66i$.

3. Results and discussion

This section presents the simulation results of different MIM plasmonic waveguides and optimisation procedure. For enhancement of SHG efficiency, four factors should be taken into account: increasing the nonlinear susceptibility $\chi^{(2)}$, reducing propagation attenuation, satisfying the phase matching condition and increasing coupling coefficients $\kappa_{1,2}$. Here, in the proposed plasmonic waveguide under consideration, these four factors can be complied through the use of asymmetric structures and by optimising the geometrical parameters of the waveguide. By using different metals at the top and bottom of the waveguide, the nonlinear susceptibility will be increased and propagation attenuation will be decreased noticeably. To increase the coupling efficiency, the voltage is applied to metals of the plasmonic waveguide. To satisfy the phase matching condition, the geometry of the structure including the ratio between two metals is optimised. All the simulations here have been calculated by using the finite-element-based commercial COMSOL software.

First, the top and bottom metals of the waveguide are optimised. Different structures including Al-LiNbO$_3$-Al, Au-LiNbO$_3$-Au, Ag-LiNbO$_3$-Ag, Al-LiNbO$_3$-Au, Al-LiNbO$_3$-Ag and Au-LiNbO$_3$-Ag have been investigated. In this situation, $d_1 = d_2$ is considered. Figure 2 shows the optical powers of FF and SHF waves, as a function of the propagation distance for a pump power of 1 W and for different structures with different metals. As is clear from the figure, the asymmetric structure has a longer distance for the FF wave and higher SHF optical power. The reason is that the space charge electric field, which couples the two modes, has an asymmetric transverse distribution. Therefore, this asymmetry is said to be favourable in the enhancement of nonlinear susceptibility and the SHG process. Moreover, for the Au-LiNbO$_3$-Ag structure, the FF optical power is more than zero even for a distance of 20 $\mu$m, as a result of reduction in propagation attenuation. The peak SHF optical power is 0.23 W and its corresponding peak position is 2.4 $\mu$m. The corresponding normalised conversion efficiency is $3.99 \times 10^6$ W$^{-1}$ cm$^{-2}$. This result is larger than that obtained in LiNbO$_3$-based PSW devices by about six orders of magnitude [12]. These results show that the proposed asymmetric structure of Au-LiNbO$_3$-Ag has the highest SHF optical power and shorter peak position in comparison with symmetric structures and other asymmetric structures. In fact, this structure has higher nonlinear susceptibility and lower propagation attenuation. Therefore, these two metals are adopted for proposed structure.

Figure 3 shows the $E_y$ distributions and frequency spectra of different structures. As shown in the figure, the peak electric field of the proposed structure is dramatically enhanced due to excitation of surface plasmon polaritons. Moreover, a second harmonic wave is generated at the centre frequency $5.68 \times 10^{14}$ Hz, and the electric field enhancement consequently led to SHG enhancement, as evidenced by the increase in the second harmonic component.

By using two different metals, the optical power of SHF increased noticeably and the peak position was shortened. In fact, the problem of small nonlinear susceptibility was solved using two different metals. To increase the power of SHF even more, the geometry of the structure could be tuned. Subsequently, the ratio between the lengths of two metals
(d1/d2) is optimised to improve the maximum power of SHF for the Au-LiNbO3-Ag structure. As mentioned earlier, the low nonlinear coupling coefficients between different frequencies, and the absorption loss of the plasmonic modes are solved using voltage and by optimising d1/d2. By adjusting the ratio between the two metals, the phase matching condition would be satisfied and the SHF could be increased noticeably. In this situation, the voltage applied to electrodes is 5 V. In the next step, the influence of the applied voltage is investigated. In Figure 4, the peak optical power of SHF and peak position of the SHG process are plotted against the d1/d2 for a FF pumping power of 1 W. As is clear from the figure, there is an optimum point for d1/d2 at which the peak optical power of SHF is maximum and its peak position is minimum. For d1/d2 of 0.4, the peak optical power of SHF is 0.44 W and its peak position is 1.2 μm. Therefore, by using two different metals and using their optimised ratio, the SHG is improved noticeably. The SHG normalised conversion efficiency as a function of d1/d2 is plotted in Figure 5. One can see

Figure 2. The optical powers of FF (top) and SHF (bottom) waves as a function of propagation distance and for different structures.
Figure 3. Electric field ($E_y$) distribution of different structures.

Figure 4. The peak optical power of SHF (top) and peak position of the SHG process (bottom) versus the $d_1/d_2$. 
Figure 5. The SHG conversion efficiency as a function of spacing.

Figure 6. The peak optical power of SHF (top) and peak position of the SHG process (bottom) versus the applied voltage.
that for the optimum value of $d1/d2$, the normalised conversion efficiency reached $2.5 \times 10^7$ $W^{-1}$ cm$^{-2}$, which is much higher than that of the conventional structure. Hence, $d1/d2$ is set to 0.4 and other parameters will be optimised for this state.

This is followed by investigation of the applied voltage. Here, the applied voltage is changed from 0 to 20 V, and for each state the SHF optical power and its peak position are calculated. Figure 6 shows the peak optical power of SHF and peak position of the SHG process as a function of applied voltage. As is clear from these results, the applied voltage influences the SHG process. Increasing the voltage caused the SHG and power consumption to increase. At a voltage of 20 V, the peak position of SHG occurred at 0.5 $\mu$m. At this voltage, the SHG conversion efficiency is as high as $5.2 \times 10^7$ $W^{-1}$ cm$^{-2}$.

Figure 7 shows the $E_y$ distributions and frequency spectra of the optimised proposed structure for an applied voltage of 5 V. A comparison of this figure with Figure 3 reveals that the peak electric field of the optimised proposed structure is dramatically enhanced.

4. Conclusions

In this study, the SHG in a novel plasmonic structure were investigated, including two different metals at the top and two different metals at the bottom. In the proposed structure, a voltage was applied on metals so as to produce a SHG electrically. Hence, the metals that defined the cavity also served as electrodes that could generate high direct current electric fields across the nonlinear material. The frequency of a fundamental wave at 458 nm was doubled and modulated in intensity by applying a moderate external voltage to the electrodes, yielding a voltage-dependent nonlinear generation with a higher coupling efficiency. By using different metals at the top and bottom of the waveguide, the nonlinear susceptibility was increased while the propagation attenuation decreased noticeably. To increase the coupling efficiency, the voltage was applied to metals of plasmonic
waveguide. To satisfy the phase matching condition, the geometry of the structure including the ratio between the two metals was optimised.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

**References**

[1] Sorger VJ, Oulton RF, Ma R-M, et al. Toward integrated plasmonic circuits. MRS Bull. 2012;37(08):728–738.
[2] Qasymeh M. Photorefractive effect in plasmonic waveguides. IEEE J Quantum Electron. 2014;50(5):327–333.
[3] Gramotnev DK, Bozhevolnyi SI. Plasmonics beyond the diffraction limit. Nat Photon. 2010;4:83–91.
[4] Schuller JA, Barnard ES, Cai W, et al. Plasmonics for extreme light concentration and manipulation. Nat Mater. 2010;9:193–204.
[5] Maier SA. Plasmonics, fundamentals and applications. New York (NY): Springer; 2007.
[6] Ozbay E. Plasmonics: merging photonics and electronics at nanoscale dimensions. Science. 2006;311(5758):189–193.
[7] Pleros N, Kriezis EE, Vyrskinos K. Optical interconnects using plasmonics and Si-photonics. IEEE J Photon. 2011;3(2):296–301.
[8] Ly Gagnon DS, Balram KC, White JS, et al. Routing and photodetection in subwavelength plasmonic slot waveguides. J Nanophoton. 2012;1(1):9–16.
[9] Goto T, Katagiri Y, Fukuda H, et al. Propagation loss measurement for surface plasmon-polariton modes at metal waveguides on semiconductor substrates. Appl Phys Lett. 2004;84:852–854.
[10] Charbonneau R, Lahoud N, Mattiussi G, et al. Demonstration of integrated optics elements based on long-ranging surface plasmon polaritons. Opt Express. 2005;13:977–984.
[11] Dionne JA, Sweatlock LA, Atwater HA. Plasmon slot waveguides: towards chip-scale propagation with subwavelength-scale localization. Phys Rev B. 2006;73:035407.
[12] Zia R, Selker MD, Catrysse PB, et al. Geometries and materials for subwavelength surface plasmon modes. J Opt Soc Am A. 2004;21:2442–2446.
[13] Miyazaki HT, Kurokawa Y. Squeezing visible light waves into a 3-nm-thick and 55-nm-long plasmon cavity. Phys Rev Lett. 2004;96:097401.
[14] Kurokawa Y, Miyazaki HT. Metal-insulator-metal plasmon nanocavities: analysis of optical properties. Phys Rev B. 2007;75:035411.
[15] Veronis G, Fan S. Bends and splitters in metal–dielectric–metal subwavelength waveguides. Appl Phys Lett. 2005;87:131102.
[16] Soltani M, Nikoufard M, Dousti M. Enhancement of second harmonic generation in metal-insulator-metal plasmonic waveguides. Plasmonics. 2017;1–5.
[17] Cazzanelli M, Bianco F, Borga E, et al. Second-harmonic generation in silicon waveguides strained by silicon nitride. Nat Mater. 2011;11(2):148–154.
[18] Levy JS, Foster MA, Gaeta AL, et al. Harmonic generation in silicon nitride ring resonators. Opt Express. 2011;19(12):11415–11421.
[19] de Oliveira REP, Lipson M, de Matos CJS. Electrically controlled silicon nitride ring resonator for quasi-phase matched second-harmonic generation. In: CLEO: science and innovations. Optical Society of America; 2012.
[20] Ning TY, Pietarinen H, Hyvärinen O, et al. Efficient second harmonic generation in silicon nitride resonant waveguide gratings. Opt Lett. 2012;37(20):4269–4271.
[21] Brongersma ML, Kik PG. Surface plasmon nanophotonics. Berlin, Heidelberg: Springer; 2007.
[22] Stockman MI. Nanoplasmonics: past, present, and glimpse into future. Opt Express. 2011;19 (22):22029–22106.
[23] Cai WS, Vasudev AP, Brongersma ML. Electrically controlled nonlinear generation of light with plasmonics. Science. 2011;333(6050):1720–1723.
[24] Davoyan AR, Shadrivov IV, Kivshar YS. Quadratic phase matching in nonlinear plasmonic nanoscale waveguides. Opt Express. 2009;17(22):20063–20068.
[25] Hasan SB, Rockstuhl C, Pertsch T, et al. Second-order nonlinear frequency conversion processes in plasmonic slot waveguides. J Opt Soc Am B. 2012;29(7):1606–1611.
[26] Ashkin A, Boyd GD, Dziedzic JM, et al. Optically-induced refractive index in homogeneities in LiNbO3 and LiTaO3. Appl Phys Lett. 1966;9(1):72–74.
[27] Yariv A. Phase conjugate optics and real-time holography. IEEE J Quantum Electron. 1978;14 (9):650–660.