Plasmonic Slot Waveguides for Third Harmonic Generation and Third-Order Parametric Down-Conversion

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Abstract The researches of third harmonic generation (THG) and third-order parametric down-conversion (TPDC) always encounter the problem of phase mismatching with such a large frequency span. Waveguides modal dispersion is an effective tool to compensate the material dispersion. However, the nonlinear polarization form of THG or TPDC is an odd function that will be almost zero if the mode field distribution contains opposite sign components. Several asymmetric plasmonic slot waveguides infiltrated in nonlinear organic material (2-[4(dimethylamino)phenyl]-3-f[4-(dimethylamino) phenyl] ethynylgbuta-1, 3-diene-1, 1, 4-tetracarbonitrile) (DDMEBT) with high third-order susceptibility are proposed. Silicon (Si) is used to fill the bottom metallic slot region to break the symmetry of the waveguide. Silver (Ag) is considered to be the metal medium due to its low loss. The needed phase-matching condition (PMC) is satisfied between the zeroth mode at fundamental frequency (FF) and the first mode at third harmonic (TH) by appropriate designing the waveguide geometrical parameters. The corresponding overlap factor for THG or TPDC is much larger than the symmetric distribution.

Key words nonlinear optics; parametric downconversion; plasmonic slot waveguides; phase matching; modal dispersion

OCIS codes 190.4410; 190.4390; 230.7370

表面等离子体沟道波导实现高效三次谐波产生与受激三光子下转换的理论研究

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摘要 三次谐波产生(THG)与三光子参量下转换(TPDC)由于频率跨度大，相位匹配条件很难实现。波导模式色散是有效补偿材料色散的工具。但是 THG 或 TPDC 的非线性极化强度为奇函数，如果模场分布存在正负分布，则奇函数几乎为 0。提出非对称等离子体沟道波导设计，沟道中引入高阶非线性有机材料(DDMEBT)。晶体硅(Si)在沟道底部以破坏波导的对称性。金属包层采用损耗较小的银(Ag)。通过设计波导几何参数，在基频(FF)零阶模式和第三次诣波(TH)一阶模式间获得了有效的相位匹配条件(PMC)，相应 THG 或 TPPC 的重叠积分远高于对称波导情况。

关键词 非线性光学；参量下转换；等离子体沟道波导；相位匹配；模式色散

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1 Introdution

Nonclassical states of light are essential to the research of quantum optics and quantum information, even in the fundamental test of quantum mechanics. Especially, entanglement states and number states are

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the important part of fundament for quantum information. Generally, photon entanglement states are photon pairs generated by spontaneous down-conversion (SPDC) by means of second order nonlinearity or four wave mixing, in which two photons are produced with various entanglement degrees of freedom, such as polarization, energy. Photon pairs can be engineered to produce high order entanglement by linear operation for the application of quantum information[1-2]. However, post-selection is required to confirm this state, which will restrict the application of quantum communications.

Direct generation of photon triplets has been a long-standing goal in quantum optics. Hubel demonstrated the first photon triplets by cascade SPDC with a very low rate[3]. Spontaneous third order parametric down-conversion (TPDC) is a one-step photon triplet generation that splits one photon into three photons. Unlike the spectral divergence in cascade SPDC, the photon triplets of spontaneous TPDC are in the same spectral region and relative higher rate than the above methods.

The TPDC is precisely a quantum process, in which the vacuum state interacts with pump photon and a photon triplet is created with the annihilation of a pump photon. TPDC can be classified into two types which are spontaneous TPDC and stimulated TPDC. The stimulated TPDC has two situations, as single photon stimulated and two-photon stimulated[4-5].

Like third harmonic generation (THG), TPDC encounters the same problems of phase-matching because of the large frequency span. Boulanger *et al*. demonstrated stimulated TPDC experiment of this process by birefringence phase matching technique in crystals, which actually is a third order different-frequency generation. Anyway, their work also gives out some valuable method of phase matching and energy correlation properties.

Here, we analyze the process of TPDC in classical nonlinear coupled wave equations. Then we propose a new scheme to get the phase-matching condition of TPDC by modal dispersion technique in a hybrid plasmonic slot waveguide.

Although TPDC is definitely a quantum process, it can also be described correctly at the stimulated condition. In this paper, we utilize the classical models to investigate the properties of TPDC in waveguides, which are the coupled wave equations for continuous conditions.

Many other nonlinear processes will have influences on TPDC, which are two-photon-absorption, free-carrier-absorption, and Raman scatterings. In addition, the loss cannot be neglected in waveguides for generality. The pump and stimulated lights are assumed in different modes of waveguides

2 Classical theory of TPDC

First, the TPDC with continuous condition is studied by the following set of nonlinear coupled-mode equations[6]:

$$\frac{\partial A_1}{\partial z} = \mathrm{i}[(I_1 \mid A_1 \mid^2 + I_2 \mid A_3 \mid^2)A_1 + I_3(A_1^*)^2 \exp(\mathrm{i}\delta\beta z)], \tag{1}$$

$$\frac{\partial A_3}{\partial z} = \mathrm{i}[(I_1 \mid A_1 \mid^2 + I_5 \mid A_3 \mid^2)A_3 + I_6(A_1)^3 \exp(\mathrm{i}\delta\beta z)], \tag{2}$$

where the nonlinear coefficients I_1, I_2, I_3, I_4, I_5, and I_6 are related to the modal overlap integrals between the fundamental and third harmonic waves and are defined as

$$I_1 = \frac{1}{12}\oiint(2 \mid \boldsymbol{F}_1 \mid^4 + \mid \boldsymbol{F}_1^2 \mid^2) \cdot n_0^2(\omega_1, \boldsymbol{r}_\perp) \cdot [k_1 n_2(\omega_1, \boldsymbol{r}_\perp)]\mathrm{d}S, \tag{3}$$

$$I_2 = \frac{1}{6}\oiint(\mid \boldsymbol{F}_1 \mid^2 \mid \boldsymbol{F}_3 \mid^2 + \mid \boldsymbol{F}_1 \cdot F_3 \mid^2 + \mid \boldsymbol{F}_1 \cdot \boldsymbol{F}_3^* \mid^2) n_0^2(\omega_1, \boldsymbol{r}_\perp) \cdot [k_1 n_2(\omega_1, \boldsymbol{r}_\perp)]\mathrm{d}S, \tag{4}$$

$$I_3=\frac{1}{4}\oiint(\boldsymbol{F}_1^*\cdot\boldsymbol{F}_3)(\boldsymbol{F}_1^*\cdot\boldsymbol{F}_1^*)n_0^2(\omega_1,r_\perp)\cdot[k_1n_2(\omega_1,r_\perp)]\mathrm{d}S, \tag{5}$$

$$I_4=\frac{1}{2}\oiint(|\boldsymbol{F}_1|^2|\boldsymbol{F}_3|^2+|\boldsymbol{F}_1\cdot\boldsymbol{F}_3|^2+|\boldsymbol{F}_1\cdot\boldsymbol{F}_3^*|^2)n_0^2(\omega_3,\boldsymbol{r}_\perp)\cdot[k_1n_2(\omega_3,\boldsymbol{r}_\perp)]\mathrm{d}S, \tag{6}$$

$$I_5=\frac{1}{4}\oiint(2|\boldsymbol{F}_1|^4+|\boldsymbol{F}_3^2|^2)\cdot n_0^2(\omega_3,\boldsymbol{r}_\perp)\cdot[k_1n_2(\omega_3,\boldsymbol{r}_\perp)]\mathrm{d}S, \tag{7}$$

$$I_6=\frac{1}{4}\oiint(\boldsymbol{F}_1^*\cdot\boldsymbol{F}_3^*)(\boldsymbol{F}_1\cdot\boldsymbol{F}_1)\cdot n_0^2(\omega_3,\boldsymbol{r}_\perp)\cdot[k_1n_2(\omega_3,\boldsymbol{r}_\perp)]\mathrm{d}S, \tag{8}$$

where $\boldsymbol{F}_1$ and $\boldsymbol{F}_3$ represent the electric mode profiles of the fundamental frequency component and the third harmonic frequency, $n_2(\omega_j,\boldsymbol{r}_\perp)$ is the nonlinear refractive index, $n_0(\omega_j,\boldsymbol{r}_\perp)$ is the linear refractive index, and $k_1=\frac{2\pi}{\lambda_1}$ stands for the wave number of the fundamental wave, λ_1 is the wavelength of the fundamental wave, integral variable S is the transverse profiles of the waveguide, I_3 is the field overlap of TPDC interaction, which contributes critically to the TPDC conversion, A_1 and A_3 refer to the slowly varying mode amplitudes of fundamental wave and third harmonic wave, respectively, $\delta\beta$ is the phase-mismatch.

3 Asymmetric plasmonic slot waveguides phase matching design

Here, we propose the asymmetric plasmonic slot waveguide (DAPSW), shown in Fig. 1. The width and height of the slot are w and h, respectively. The considered Kerr-type DDMEBT with a third-order susceptibility $\chi^{(3)}=2\times10^{-19}$ m^2/V^2 and a refractive index $n=1.8$ at the wavelength of 1550 nm was infiltrated into the metallic slot. Metal, considered to excite the surface plasmon polaritons (SPPs), is silver due to its relatively low loss and widely application in the plasmonic devices. Using this DAPSW, strong third order optical nonlinear interaction and efficient phase-matched THG can be achieved between the fundamental mode at the fundamental frequency (FF) and the first order mode at the third harmonic frequency (THF).

By solving the rigorous modal eigenvalue equations with the finite-element-based COMSOL software, we analyze the guided modes for different waveguide slot widths. In the Fig. 2(a), we calculate the effective indices of the two guided modes as a function of the slot widths for a certain slot height $h=40$ nm. The black and blue lines represent the calculated effective indices for modes at FF ($\lambda_1=3600$ nm) and THF ($\lambda_3=1200$ nm). The cross point in the figure denotes that the phase matching condition is satisfied when the slot width is set to be 509 nm. At this condition, the effective mode refractive indices of the FF and THF

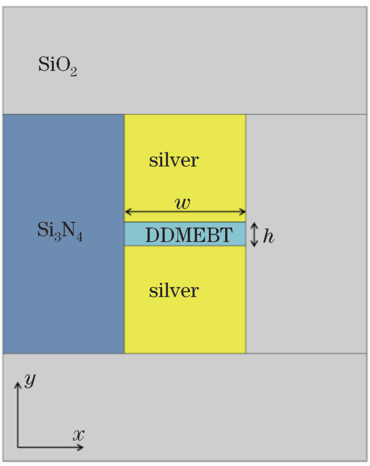

Fig. 1 Schematic cross-section of the proposed metal-organic-metal plasmonic slot waveguide

are 2.446796+0.01669i and 2.446778+0.009118i, respectively. The TPDC overlap, $I_3=21.494\ \mu m^{-2}$, much smaller than other factors as self-phase modulation (SPM) and cross-phase modulation (XPM).

4 Double asymmetric plasmonic slot waveguides phase matching design

Figure 3 shows the cross-section view of the proposed DAPSW. The widths and heights of the slot are w_1 (slot width), w_2 (silicon slot width), h_1 (slot height) and h_2 (silicon slot height), respectively. Fig. 4(a) and 4(b) depict the corresponding two dimensional dominant electric field component Ex distributions for the zeroth mode at FF and the first mode at TH under the PMC. It can be found that, the electric components of the zeroth mode at FF are all positive while the first mode at TH preserves both positive and negative parts. We also plot the one dimensional electric field distributions along a y cutline at $x=0$ [as the dash lines shown in Fig. 4(a) and 4(b)] in Fig. 4(c). Similarly, Fig. 4(d) illustrates the one dimensional electric field distributions along a x cutline at $y=0$. Two different nonlinear materials are integrated into the metallic slot region and the silicon slot is set to be wider than the organic material slot, while the cladding of the DAPSW is different from the substrate. Due to the double-asymmetric structure, the entire negative part of the 1-st mode at pump distributes into the silicon slot region, which greatly reduces the corresponding counteractive effect to the pump-harmonic modal overlap. Furthermore, the enhancement of the negative part of the higher-order mode at pump is significantly reduced compared with the positive part, which further enhances the modal overlap to the range of 4000 μm^{-2}.

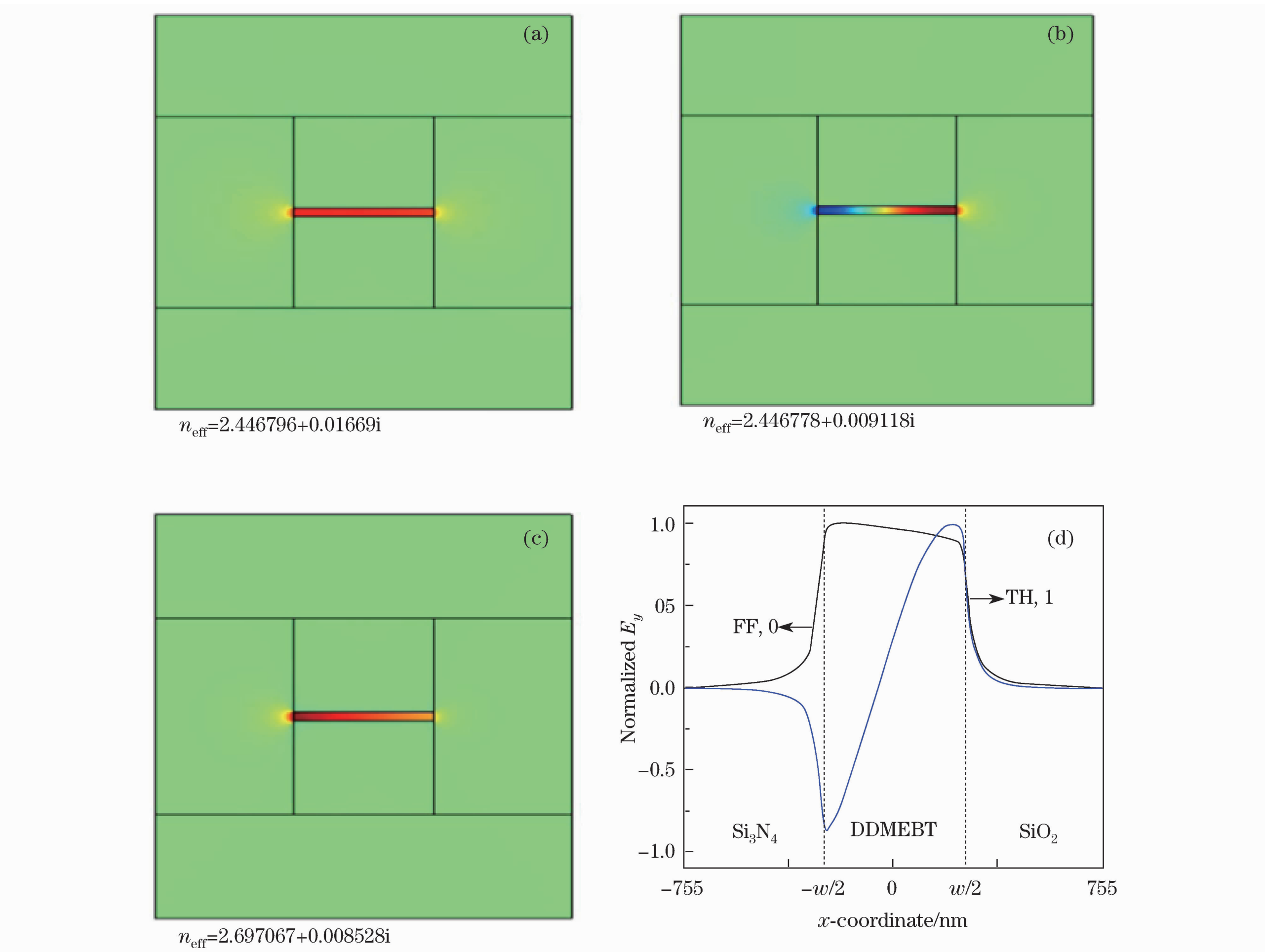

Fig. 2 (a) 0-th mode at FF; (b) 1-st mode at THF; (c) 0-th mode at THF; (d) normalized electric field E_y distributions along $y=0$

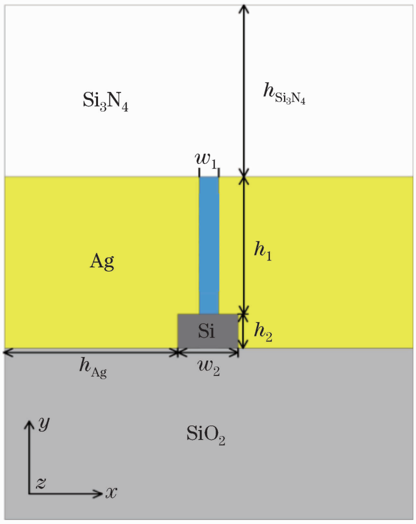

Fig. 3 Cross-section view of the proposed DAPSW

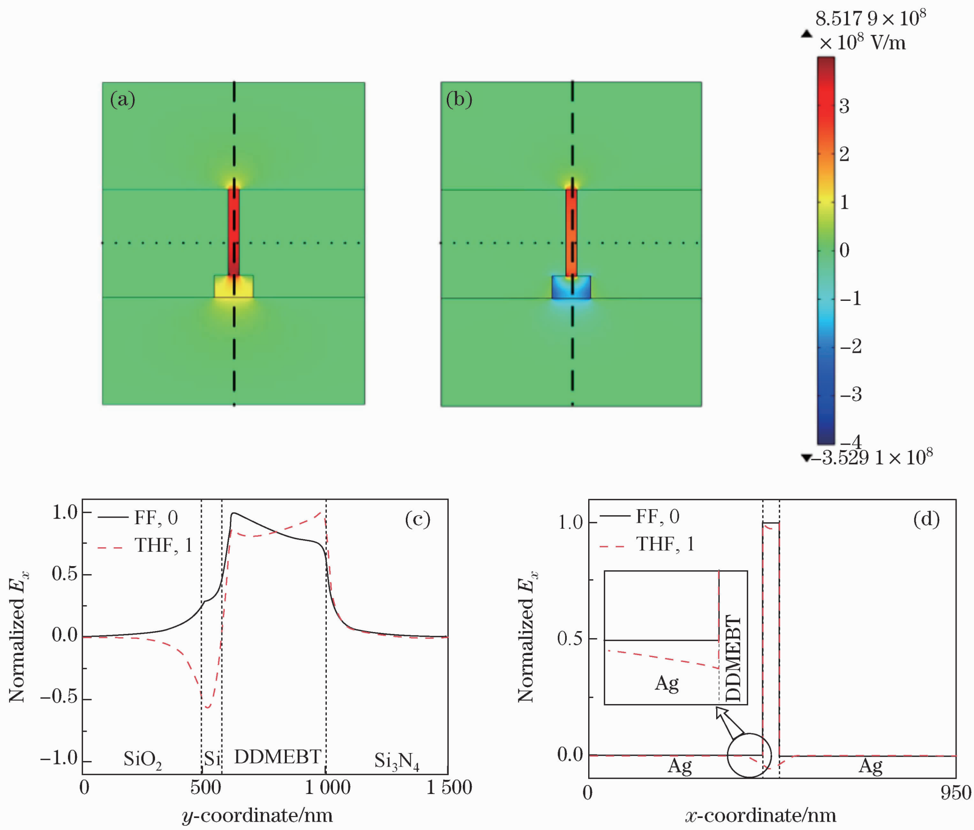

Fig. 4 Major 2D E_x profiles of (a) 0-th mode at FF and (b) 1-st mode at THF; 1D E_x distributions of FF and THF at a cutline of (c) $x=0$ as the vertical dash lines plotted in Fig. (a) and (b), and (d) $y=0$ as the horizontal dot lines plotted in Fig. (a) and (b) ('0' stands for the 0-th mode, '1' stands for the 1-st mode)

5 Conclusion

The asymmetric plasmonic slot waveguides are proposed to get the phase matching condition for TPDC. The double asymmetric structure can enhance the TPDC process greatly.

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