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Efficient Generation $1.5THz$ Pulses Using $DASC/GaAs$ and $DASB/GaAs$ Multilayer Structures via Difference Frequency Generation $1.5\ \mu m$ Femtosecond Laser Pulses

Maryam Kashani and Hamid Reza Zangeneh ⋆

Abstract

We present a study of high efficient terahertz (THz) wave generation in $DASB/GaAs$, $DASC/GaAs$ and $DAST/GaAs$ multilayer structures at $1.5THz$ via difference frequency generation (DFG) process with $1.5\mu m$ femtosecond laser pulses. We also compare the conversion efficiency in proposed structures with $DASB/SiO_2, DASC/SiO_2$ and $DAST/SiO_2$ multilayer structures and bulk crystals $DASB, DASC$ and $DAST$. These structures compensate for phase mismatching in bulk crystals $DASB, DASC$ and $DAST$ and increase conversion efficiency from 10^{-5} in bulk organic crystals up to 10^{-3} in multilayer structures. We show that $DASC/GaAs$ is the best structure to generate $1.5THz$ waves.

Keywords: terahertz waves (THz), difference frequency generation (DFG), nonlinear susceptibility, multilayer structure, organic crystals.

2020 Mathematics Subject Classification: 78A50, 78A60, 78M15.

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1. Introduction

The tunable coherent terahertz (THz) sources are the important devices for imaging and time-domain spectroscopy (TDS) applications [13]. Recently, high power ultra-short waves with average frequency below $1.5THz$ have been generated via optical rectification of femtosecond laser pulses with tilted pulse fronts in lithium niobate ($LiNbO_3$) crystal. But attempts for utilizing this method to generate laser pulses with frequencies higher than $1.5THz$ lead to decreases in THz efficiency owing to increases in THz absorption in this material [11]. High efficiency pump-to-THz conversion to generate the frequency range between 1 till $10THz$, using DFG in organic salt crystals 4-dimethylamino-N'-methyl-4′-stilbazolium p-chlorobenzenesulfonate ($DASC$), 4-dimethylamino-N'-methyl-4′-stilbazolium p-bromobenzenesulfonate ($DASB$), 4-dimethylamino-N'-methyl-4′-stilbazolium tosylate ($DAST$), 2-{3-(4-hydroxystyryl)-5, 5-dimethylcyclohex-2-enylidene} malononitrile ($OH1$), 4-N, N-dimethylamino-4′-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate ($DSTMS$) have been established [6, 12, 13]. Multilayer structure $DAST/SiO_2$ has been proposed before to generate quasi-single-cycle pulses with average frequency 6THz by DFG femtosecond 800 nm laser pulses [10]. Here, we consider generation of $1.5THz$ pulses via DFG femtosecond $1.5\mu m$ laser pump in multilayer structures $DASB/GaAs$, $DASC/GaAs$, $DAST/GaAs$, $DASB/SiO_2$, $DASC/SiO_2$ and $DAST/SiO_2$ and compare their THz efficiencies with each other. The reason for utilizing pump laser with an average wavelength of $1.5\mu m$ for generating THz pulses is low absorption of pump waves and almost perfect phase matching in these materials.

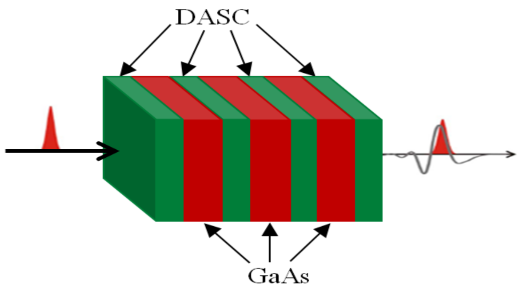

Figure 1: Multilayer structure of DASC/GaAs.

2. Theory and Modeling

Many of stilbazulium salts have been established as high nonlinear optical crystals. Organic crystal $DAST$ has large second-order optical nonlinearity (d_{11} =

$290pm/V at 1542nm$ [13]). For this reason, it is a very attractive material in nonlinear research and its applications [7]. Having high second-order susceptibility and velocity matching between pump waves and THz pulses in crystals are important factors for selecting crystals as THz sources [3]. But another important factors to increase the figure of merit (FM) for efficiently generating THz waves is low absorption in THz region [8]. Hence, some of stilbazulium salts with low THz absorption coefficient compared to $DAST$ are more attractive as THz sources. $DASC$ and $DASB$ are $DAST$'s derivatives with an ability to generate THz waves more efficiently than DAST [7]. $DASB$, $DASC$ and $DAST$ have approximately the same value of second - order optical nonlinearity as a result of similar structure. But, $DASC$ and $DASB$ have lower THz absorption coefficients than $DAST$. Thus, they have higher transmittance and THz output power compared with $DAST$. Moreover, the FMs of $DASC$ and $DASB$ are nearly twice that of $DAST$ [7, 8, 9]. The absorption coefficients of $DAST$ and $DASB$ crystals at $1.5THz$ are equal to $115cm^{-1}$ and $80cm^{-1}$, respectively [2, 7], whereas the absorption coefficient of $DASC$ at THz region is equal to $10cm^{-1}$ [8, 9]; therefore, it is expected that the FM of $DASC$ is higher than that of $DAST$ and $DASB$. Figure 2 shows that $DASC$, $DASB$ and $DAST$ crystals have the same value of THz refractive index.

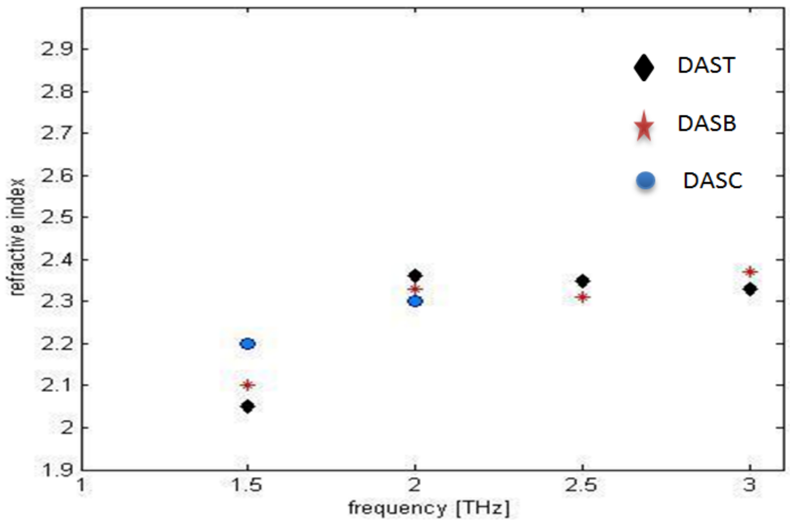

Figure 2: THz refractive index of $DASC$, $DASB$ and $DAST$ crystals.

These curves have been calculated using:

$$T = \frac{16n_{THz}^2}{(n_{THz}+1)^4} e^{-\alpha_{THz} z},$$

where T stands for transmittance, α_{THz} and n_{THz} are absorption coefficient and refractive index in THz region respectively, and z is the crystal thickness [7, 8, 9]. We can estimate that due to having the same molecule structure and symmetry versus axis, the group refractive indexes in $DASC$, $DASB$ and $DAST$ crystals have approximately the same value. We have used these crystals in multilayer structures $DASB/GaAs$, $DASC/GaAs$, $DAST/GaAs$, $DASB/SiO_2$, $DASC/SiO_2$ and $DAST/SiO_2$ to generate $1.5THz$ waves by DFG of two pump pulses with an average wavelength of $1.5\mu m$. Multilayer structures have been designed to compensate for phase mismatching that accompanies the generation of THz pulses in $DASC$, $DASB$, $DAST$ [10]. The group refractive index of $DAST$ at 1.5 μm ($n_g^{DAST} = 2.26$[1]) is higher than THz refractive index at $1.5THz$ ($n_T^{DAST} = 2$[2]) whereas in GaAs [4]:

$$n_g^{GaAs}(1.5\mu m) = 3.52 < n_T^{GaAs}(1.5THz) = 3.6.$$

In quartz in the same wavelength region for ordinary ray we have [4, 5]:

$$n_g^{0-quartz}(1.5\mu m) = 1.46 < n_T^{quartz}(1.5THz) = 2.1 - 2.2.$$

In fact, the aim of using these structures is to generate near- single- cycle pulses [10]. We use $GaAs$ and SiO_2 layers to remove phase mismatching that appears in organic crystals. $GaAs$ is a nonlinear media ($d_{14}^{GaAs} = 46.1pm/V$ [14]), therefore this crystal acts as a THz source and in addition to compensating for phase mismatching, it can help to generate and increase intensity of THz waves and lead to increases in efficiency.

$$-\nabla^2 \tilde{E}_T + \frac{\epsilon^{(1)}}{c^2}\frac{\partial^2 \tilde{E}_T}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2 \tilde{P}_T^{NL}}{\partial t^2}, \tag{1}$$

where $\tilde{E}_T$ and $\tilde{P}_T^{NL}$ are the electric field of THz and nonlinear polarization of the THz wave, respectively, and $\epsilon^{(1)}$ is the dielectric constant. $\tilde{E}_T$ and $\tilde{P}_T^{NL}$ are described by the following equations:

$$\tilde{P}_T = P_T e^{-i\omega_T t}, \tag{2}$$

$$P_T = 4d_{eff} E_1 E_2^{\star}, \tag{3}$$

$$E_1 = A_{p_1} e^{-ik_1 z} e^{\frac{\alpha_1}{2} z}, \tag{4}$$

$$E_2^{\star} = A_{p_2}^{\star} e^{ik_2 z} e^{\frac{\alpha_2}{2} z}. \tag{5}$$

By substituting Equations $(2-5)$ in (1) and solving differential equation, we derive the equation that describes amplitude of THz waves [15]:

$$\frac{dA_T}{dz} = -\frac{\alpha_T}{2}A_T - i\frac{8\pi\omega_T}{n_T c} d_{eff} A_{p_1} A_{p_2}^{\star} e^{-i\Delta kz}, \tag{6}$$

where A_T , A_{p_1} and A_{p_2} are imaginary amplitudes of THz and laser pump waves respectively. Also α_T and n_T are absorption coefficient and refractive index of THz wave at ω_T frequency respectively. d_{eff} is the coupling constant; and $\Delta k = k_1 - k_2 - k_T$ propagation vector mismatching in organic crystals. By solving Equation (6):

$$A_T = -i\frac{8\pi\omega_T d_{eff} A_{p_1} A^\star_{p_2}}{n_T c(\alpha_T/2 - i\Delta k)} e^{-i\Delta k L} + A_i e^{-\frac{\alpha_T}{2}L}, \tag{7}$$

where A_i is integration constant for one layer. By solving Equation (6) for the first layer we have

$$A_T = -i\frac{8\pi\omega_T d_{eff} A_{p_1} A^\star_{p_2}}{n_T c(\alpha_T/2 - i\Delta k)} e^{-i\Delta k L} + i\frac{8\pi\omega_T d_{eff} A_{p_1} A^\star_{p_2}}{n_T c(\alpha_T/2 - i\Delta k)} e^{-\frac{\alpha_T}{2}L}, \tag{8}$$

and for the second layer

$$A_T = i\frac{8\pi\omega_T d_{eff} A_{p_1} A^\star_{p_2}}{n_T c(\alpha_T/2 - i\Delta k)} (e^{-\frac{\alpha_T}{2}L_1} - e^{-i\Delta k L_1}) e^{-\alpha_q(L_1-L_2)}/2. \tag{9}$$

By using

$$\eta = \frac{P_{THz}(L)}{P_{optical}(0)}, \tag{10}$$

for THz wave we have

$$\eta = \frac{128\pi^3\omega_T^2 d_{eff}^2 I}{n_1 n_2 n_T^2 c^3(\frac{\alpha_T^2}{4} + \Delta k^2)}[1 - e^{(-\frac{\alpha_T}{2} - i\Delta k)L} - e^{(-\frac{\alpha_T}{2} + i\Delta k)L} + e^{-\alpha_T L}], \tag{11}$$

where I is laser pump intensity, n_1 and $n_2$are the refractive indexes of first and second laser pumps, respectively. Using equation below, we can estimate thickness ratios of layers to obtain full phase-mismatch compensation

$$t_g^{organic\ layer} + t_g^{quartz\ layer} = t_{THz}^{organic\ layer} + t_{THz}^{quartz\ layer}, \tag{12}$$

where t_g and t_{THz} are times of propagation of optical and THz waves in organic and quartz layers, respectively.

$$\frac{1}{v_g^{organic\ crystal}} + \frac{\beta}{v_g^{quartz}} = \frac{1}{v_{THz}^{organic\ crystal}} + \frac{\beta}{v_{THz}^{quartz}}, \tag{13}$$

$$\beta = \frac{v_{THz}^{quartz} v_g^{quartz}(v_{THz}^{organic\ layer} + v_g^{organic\ layer})}{v_{THz}^{organic\ layer} v_g^{organic\ layer}(v_g^{quartz} - v_{THz}^{quartz})}, \tag{14}$$

where v_g and v_{THz} are group velocity in quartz layer and THz velocity in organic crystal layer, respectively; and β is the thickness ratio of quartz layer to organic crystal layer [15].

3. Results and Discussion

All of multilayer structures consist of 5 organic and 4 inorganic layers. Reflection from boundaries and phase matching are major factors in determining the number of layers and to obtain maximal efficiency. Gradually decreasing the thickness of organic and inorganic layers causes the best phase matching between optical and the THz waves and hence grater conversion efficiency. In fact, we can prevent THz absorption in deference layers by a smooth decrease in thickness of layer. The thickness values of layers in the structures under study have been summarized in Figure 5. In multilayer structures $DASC/GaAs$, $DASB/GaAs$ and $DAST/GaAs$ that have been proposed for generating $1.5THz$ waves, $GaAs$ layers for obtaining complete phase matching have to be thicker than $DASC$, $DASB$ and $DAST$ layers by a factor of 1.1; and the thickness ratios of layers in $DASC/SiO_2$ and $DASB/SiO_2$ structures which were proposed to generate $1.5THz$ waves are 1.1. But in the $DAST/SiO_2$ at this frequency, $DAST$ and SiO_2 layers have the same thickness. Reflection of optical waves and THz waves from common surface in $DASC$, $DASB$ and $DAST/GaAs$ structures is 1.6% and 8.41% respectively, and reflection from the air and organic crystals is 5.07%. These same values in $DASC$, $DASB$ and $DAST/SiO_2$ structure are 1.9% and 0.23%, respectively.

THz generation efficiency as a function of distance in bulk $DASC$, $DASB$ and $DAST$ crystals and multilayer structures have been shown in Figures 3 and 4.

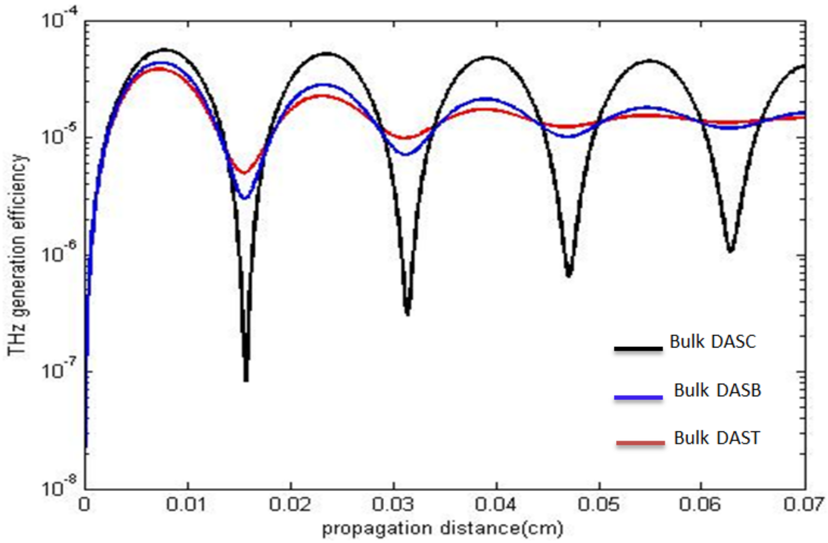

Figure 3: THz conversion efficiency in bulk $DASC$,$DASB$ and $DAST$ at $1.5THz$.

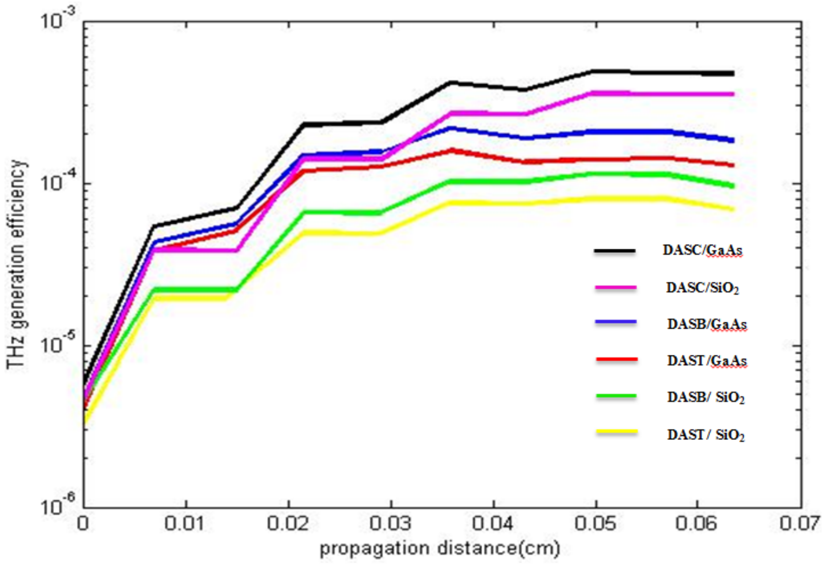

Figure 4: THz efficiency in multilayer Str. $DASC/GaAs$, $DASB/GaAs$, $DAST/GaAs$, $DASB/SiO_2$, $DASC/SiO_2$ and $DAST/SiO_2$ at $1.5THz$.

We can see that bulk $DASC$ shows higher efficiency than $DASB$ and $DAST$ due to its lower THz absorption. Figure 4 shows that $DASC/GaAs$ multilayer structure has a higher efficiency than all other structures, because $GaAs$ is a nonlinear medium; hence it is known as a source of THz waves and therefore increasing optical-to-THz conversion efficiency, but SiO_2 is a linear medium and only fixes the conversion efficiency.

4. Conclusion

In this numerical study, we have presented multilayer structures for generating single-cycle $1.5THz$ pulses via DFG process at 1.5 μm femtosecond optical waves because these structures are more efficient than bulk crystals. In this wavelength, phase matching is almost perfect and absorption of pump waves is low. $GaAs$ and SiO_2 layers prevent decrease in efficiency in multilayer structures in spite of phase mismatching in bulk organic crystals. $GaAs$ is a source of THz generation therefore it increases laser-to-THz energy conversion efficiency whereas SiO_2 fixes conversion efficiency and also $DASC$ crystal has lower absorption at $1.5THz$ than $DASB$ and $DAST$ crystals. Hence among all of alternating structures, conversion efficiency of $DASC/GaAs$ structure is higher than other multilayer structures. In addition multilayer structures are a new trend to generate high efficient THz pulses with an average frequency below 2 THz which was difficult to obtain previously.

DASB/SiO_2 at 1.5THz		
Layer number	Material	Thickness (μm)
1	DASB	70
2	SiO_2	80
3	DASB	65.7
4	SiO_2	75.48
5	DASB	67
6	SiO_2	73.7
7	DASB	66
8	SiO_2	72.6
9	DASB	65

DASC/SiO_2 at 1.5THz		
Layer number	Material	Thickness (μm)
1	DASC	70
2	SiO_2	80
3	DASC	65.7
4	SiO_2	75.48
5	DASC	67
6	SiO_2	73.7
7	DASC	66
8	SiO_2	72.6
9	DASC	65

DAST/SiO_2 at 1.5THz		
Layer number	Material	Thickness (μm)
1	DAST	70
2	SiO_2	70
3	DAST	75
4	SiO_2	76
5	DAST	67
6	SiO_2	67
7	DAST	66
8	SiO_2	66
9	DAST	65

TBC(DAST, DASB, DASC)/GaAs at 1.5THz		
Layer number	Material	Thickness (μm)
1	TBC	70
2	GaAs	80
3	TBC	65.7
4	GaAs	75.48
5	TBC	67
6	GaAs	73.7
7	TBC	66
8	GaAs	72.6
9	TBC	65

Figure 5: Thicknesses of layers in studied structures at $1.5THz$.

Conflicts of Interest. The authors declare that there are no conflicts of interest regarding the publication of this article.

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