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Generation of High Efficient Quasi-Single-Cycle 3 and 6 THz Pulses using Multilayer Structures $OH1/SiO_2$ and $DSTMS/SiO_2$

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Abstract

We propose that high efficient terahertz (THz) multilayer structures are composed of $DSTMS/SiO_2$ and $OH1/SiO_2$ at 3 and 6 THz frequencies. We show that the efficiencies of these structures are higher than $DAST/SiO2$ structure in both of 3 and 6 THz frequencies. $OH1/SiO_2$ structure at 6 THz has an efficiency as large as 10^{-1}; at 3 THz frequency, $DSTMS/SiO_2$ structure has an efficiency as large as 10^{-2}. Meanwhile bulk $OH1$ has an efficiency as large as 10^{-3} at 3 THz due to perfect phase matching whose efficiency is lower than $DSTMS/SiO_2$ structure. We also show that other structures, namely $DSTMS/ZnTe$ at 3 THz and $DAST/GaP$ at 8 THz, have low efficiency, so they are not suitable as THz sources.

Keywords: Terahertz waves (THz), difference frequency generation (DFG), non-linear susceptibility, multilayer structure, organic crystals.

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1. Introduction

During the past decade, high-energy ultra-short terahertz (THz) pulses with average frequency below 2 THz have been obtained at large accelerator facilities

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or through optical rectification (OR) of femtosecond laser pulses with a nonlinear medium. Due to their simplicity and broad band, THz generated waves are of foremost importance [8]. Nonlinear optical effects such as difference frequency generation (DFG) or optical rectification (OR) are widely used for generating coherent THz radiation. Since appropriate source materials have a high second order nonlinear optical susceptibility $\chi^{(2)}$ and a low refractive index in the THz frequency range, velocity-matched conversion from optical wave to THz wave occurs [11]. Using DFG, high power (up to 0.5GW) few-cycle pulses with tunable frequencies from 10 to 72 THz have been generated. Despite this progress, however, high-power sources in the ever important frequency range of 3-9 THz are still nonexistent. High-energy ultrashort THz pulses in this range are desirable for many scientific applications, such as nonlinear probing of the fundamental lattice vibration of polar crystals and two-dimensional THz spectroscopy of the nonlinear vibrational response of water [8]. Organic noncentrosymmetric materials are highly sought after for applications in nonlinear optics. These materials provide a wide range of design possibilities to the scientists. Due to an almost-complete electronic origin of the nonlinearity, they are well suited for future high-speed devices [7]. Considerable efforts have been made in well-established design strategies that lead to large microscopic molecular optical nonlinearities of molecules based on a highly extended π-conjugated bridge between electron donor and acceptor groups [5]. High pump-to-THz conversion efficiency to generate the frequency range between 1 and 10 THz, using DFG in organic salt crystals 2-{3-(4-hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene} malononitrile (OH1), 4-N, N-dimethylamino-$4' - N'$-methyl-stilbazolium 2, 4, 6-trimethylbenzenesulfonate (DSTMS) and 4-N,N-dimethylamino-$4' - N'$-methyl-stilbazolium tosylate (DAST) have been established. Reaching large conversion efficiency in many organic crystals needs femtosecond laser pulses [10]. As reported 12 years ago, Laser-to-THz energy conversion efficiency has been increased from 10^{-8}, up to the recently achieved value of 10^{-4}. It was also proposed to use multilayer structure $DAST/SiO_2$ to efficiently generate nearly single-cycle pulses with an average frequency of 6 THz, generated via difference frequency generation at $800nm$ using femtosecond laser pulses [8].

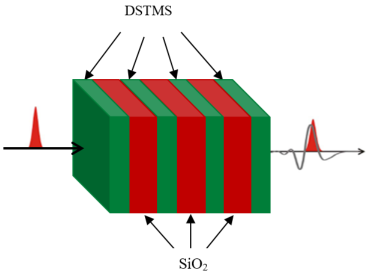

Figure 1: Multilayer structure of $DSTMS/SiO_2$.

Here, we propose $DSTMS/SiO_2$ and $OH1/SiO_2$ multilayer structure for efficient generation of nearly single-cycle 6 and 3 THz pulses and compare THz efficiencies of all the structures.

2. Theory and Modeling

Organic crystals are used as a source for generating and detecting terahertz waves using OR and DFG. Materials used to generate THz waves should have two properties. First, they must have a high second order susceptibility, and second, velocity matching in crystals for THz generation and detection. These properties are attainable in the organic crystals with low dielectric constant [4]. $DAST$ is currently the only commercially available material with very large second order nonlinear properties $d_{111} = 210pm/V$ at wave length $1.9\mu m$ with low dielectric constant ($\epsilon = 5.2$) among organic crystals. Cultivation of $DAST$ with high optical properties is still a challenge [7] and many research groups are interested in investigating the growth of bulk and thin types of this crystal [14].

Recently, a new and promising type of organic crystals that shows desirable properties is $DSTMS$ [7]. $DSTMS$ chemical structure is similar to $DAST$ chemical structure with very good roughness ($\lambda/100$ at THz frequencies), i.e. it does not need surface polishing. We use x direction in $DSTMS$ due to the largeness of $\chi^{(2)}_{111}$ in this direction [11]. Another optical organic material in high technology is $OH1$, with highly desirable, specific and favorable electro-optical nonlinear properties. These crystals with high nonlinear susceptibilities suitable for many applications are very promising in nonlinear optics [4]. Compared to $DSTMS$ and $DAST$, $OH1$ has larger figure of merit (FM). The absorption coefficient of THz waves in frequencies between $0.3 - 2.5THz$ in $OH1$ is very low and in frequency range of $0.7 - 1THz$, the absorption coefficient is lower than $0.2mm^{-1}$ i.e. similar to absorption coefficient in $ZnTe$ and $GaAs$ but in contrast with $DAST$ in the same frequency range. $OH1$ molecule has a great magnetic dipole, $\mu_g = 3.44\times10^{-29}Cm$, and its π- electron structure is so wide and long that it results in high polarization capability. Table 1 shows FM of $OH1$, $DSTMS$, $DAST$, $GaAs$ and $ZnTe$ [1].

Table 1: Comparison of different materials in terms of eligibility criteria, absorption and electro-optical coefficients [1, 14].

Material	$\lambda(\mu m)$	n_o	n_g	$r(pm/V)$	$FM(pm/V)^2$	$\alpha(mm)^{-1}$
OH1	1.3	2.16	2.33	52.0	5300	0.2
DSTMS	1.9	2.08	2.19	49.0	6100	0.9
DAST	1.5	2.13	2.26	47.0	4200	3-5
ZnTe	0.8	2.85	3.23	4.0	370	0.1
GaAs	1.4	3.40	3.61	1.3	86	0.3

In this paper, we propose two multilayer structures for increasing the efficiency generation of THz waves in $OH1$ and $DSTMS$. Using multilayer structure of $DAST/SiO_2$ to generate effective pulses i.e. nearly-single-cycle with average frequency of 6 THz by optical rectification femtosecond laser $800nm$ has been proposed [8].

Proposed $OH1/SiO_2$ and $DSTMS/SiO_2$ structure generate almost single-cycle pulses with frequencies at 3 and 6 THz, and $DAST/SiO_2$ structure for frequency 3 THz as well. Reason of using such a structure is to remove phase mismatching. In quartz (SiO_2) crystal, phase velocity of THz wave in a wide range of frequency is lower than group velocity of laser pulses with $800nm$ wavelength.

Tables 2 and 3 show refractive indexes, absorption coefficients and nonlinear susceptibilities of different crystals at $800nm$.

Table 2: Absorption coefficient and the nonlinear susceptibilities for different crystals at 800 nm and refractive indices at frequencies 3 and 6 THz.

Crystal	d_{eff} (pm/v)	n_t (3 THz)	n_t (6 THz)	n_g (800 nm)	$\alpha(cm)^{-1}$ (3 THz)	$\alpha(cm)^{-1}$ (6 THz)	Ref.
DSTMS	670.35	2.20	2.20	3.40	50.0	9.66	[7, 11, 13, 14]
OH1	600.42	2.10	2.23	2.42	280.0	11.14	[1, 2, 4, 13]
DAST	615.00	2.36	2.36	3.38	100.0	140.00	[3, 8]
SiO_2	Linear Response	2.13	2.20	1.55	1.6	5.00	[6, 8]
ZnTe	68.5	3.42	$3.27i$	3.24	30.0	301.00	[6]

The measured refractive indexes for $DSTMS$ and $OH1$ at 6 THz frequency have been obtained by equation:

$$l_c = \frac{c}{2\nu_{THz}[n_t - n_g(\lambda_p)]};$$

where l_c is coherence length, n_t and n_g are THz refractive index and the group refractive index respectively, c is the speed of light in vacuum and ν_{THz} is the THz frequency and λ_p is the optical wavelength of pump [13].

For $DSTMS$ at $\lambda_p = 1.5\mu m$ and 6 THz the coherence length and the group refractive index are $l_c = 1mm$, $n_g = 2.3$ respectively and for $OH1$ at $\lambda_p = 1.5\mu m$ and 6 THz, $l_c = 0.4mm$, $n_g = 2.25$.

Table 3: Absorption coefficient and the nonlinear susceptibilities for different crystals at 800 nm and refractive indices at frequencies 3 and 8 THz

Crystal	d_{eff} (pm/v)	n_t (3 THz)	n_t (8 THz)	n_g (800 nm)	$\alpha(cm)^{-1}$ (8 THz)	Ref.
GaP	24.8	3.36	3.68	3.55	39.43	[12]
DAST	615.0	2.36	2.36	3.38	200.00	[3, 8]

In this study, using difference frequency generation (DFG) process between $793nm$ and $806nm$ pulses, we compute efficiency at 6 THz. Meanwhile by using

797nm and 803nm pulses we compute efficiency at 3 THz, and then compare the results of 6 and 3 THz waves.

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

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:

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

$$P_T = 4d_{\text{eff}} E_1 E_2^* \tag{3}$$

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

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

By substituting equations (2)-(5) in (1) and solving differential equation, for amplitude of THz waves we derive:

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

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 $\Delta k = k_1 - k_2 - k_T$ is the propagation vector mismatching in organic crystals. By solving equation (6):

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

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

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

and for the second layer:

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

by use of:

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

For THz wave we have:

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

where I is laser pump intensity, n_1 and n_2 are the refractive indices of first and second laser pumps, respectively. Using quartz layers in this structure compensates the phase mismatching between optical and THz waves. Using OR or DFG processes for $800nm$ pulses to generate waves with average frequency larger than 1 THz is not effective [8]. So, multilayer structure is a method for solving this problem. SiO_2 layers compensate for group velocity mismatching of optical and THz waves at $800nm$. So multilayer layers of SiO_2 and $DAST$ have been used [8]. Other alternating structures are $OH1/SiO_2$ and $DSTMS/SiO_2$ with suitable thickness in order to avoid phase mismatching and to obtain greater efficiencies. In alternating structure for prefect compensation of phase mismatching, quartz layers have to be thicker than $OH1$, $DSTMS$ and $DAST$ layers.

Using equation below, we can estimate thickness ratios of layers to obtain full phase-mismatch compensation:

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

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

$$\beta = \frac{v_{THz}^{\text{quartz}} v_g^{\text{quartz}} (v_{THz}^{\text{organic layer}} + v_g^{\text{organic layer}})}{v_{THz}^{\text{organic layer}} v_g^{\text{organic layer}} (v_g^{\text{quartz}} - v_{THz}^{\text{quartz}})} \tag{13}$$

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.

For $DSTMS/SiO_2$ structure, β is approximately from 1.3 up to 2.18 at 3 THz. The selected value is 1.5 to achieve higher efficiency; for the same structure, this ratio is 2.6 at 6 THz frequency; in $OH1/SiO_2$ structure at 6 THz frequency the thickness ratio is from 1 up to 1.2. Similarly this ratio for quartz layers for the $DAST$ layers in $DAST/SiO_2$ varies from 1.35 to 1.85 at 3 and 6 THz. Reflection from boundaries is a major factor in calculating the efficiency and fixing layers number; then for optical waves in $DSTMS/SiO_2$ structure, reflection from common surface is 4.5% and reflection from the air and $DSTMS$ is 11.4%. Due to small differences between refractive indexes of $DSTMS$ and quartz (Table 2) at 3 THz wave, reflection from common surface is 0.06% that this value at 6 THz vanishes. In $OH1/SiO_2$ structure, reflection from common surface between $OH1$ and SiO_2 is 1.37% and reflection from the air and $OH1$ is 5.37%. On the other hand, for waves at 6 THz frequency this value reaches 0.005% which means THz wave propagates from $OH1$ to SiO_2 without damping. In $DAST/SiO_2$ structure for optical waves at 6 THz reflection from boundary between air and $DAST$ is 16.5% and reflection from common surface $DAST$ and SiO_2 is 4.5% [8] and the same values in this structure at 3 THz are 16.5% and 4.5% respectively. By the way, THz reflection from common surface between $DAST$ and SiO_2 is 0.1% at 3 THz. So we can approximate the number of layers in structures $OH1/SiO_2$, $DSTMS/SiO_2$ and $DAST/SiO_2$ i.e. the number of layers for $OH1$, $DSTMS$ and $DAST$ is 11 layers, and 10 layers for SiO_2 in three structures for both 3 and 6 THz frequencies. Calculations show that the greatest efficiency can be obtained by using monotonous decreasing in thickness of layers in every structure [8].

The thickness values of layers in the structures under study are summarized in Tables 4 and 5.

Table 4: Thicknesses of layers in studied structures of $DAST/SiO_2$ at 3 and 6 THz.

$DAST/SiO_2$(at 3THz)

Layer number	Material	Thickness(μm)
1	$DAST$	10
2	SiO_2	15
3	$DAST$	9
4	SiO_2	13.5
5	$DAST$	9
6	SiO_2	13.5
7	$DAST$	8
8	SiO_2	12
9	$DAST$	8
10	SiO_2	12
11	$DAST$	8
12	SiO_2	12
13	$DAST$	8
14	SiO_2	12
15	$DAST$	7
16	SiO_2	10.5
17	$DAST$	6.3
18	SiO_2	9.5
19	$DAST$	5.5
20	SiO_2	8
21	$DAST$	5.5

$DAST/SiO_2$(at 6THz).

Layer number	Material	Thickness(μm)
1	$DAST$	21
2	SiO_2	34
3	$DAST$	19.8
4	SiO_2	29.7
5	$DAST$	18
6	SiO_2	27
7	$DAST$	17
8	SiO_2	25.5
9	$DAST$	16.5
10	SiO_2	24.7
11	$DAST$	16
12	SiO_2	24
13	$DAST$	15
14	SiO_2	22.5
15	$DAST$	15
16	SiO_2	22.5
17	$DAST$	13.7
18	SiO_2	19.2
19	$DAST$	12
20	SiO_2	16.5
21	$DAST$	12

The thickness values of organic layers are determined to reach maximal efficiency, in fact the organic layers are computed as where the conversion efficiency starts decreasing and the thickness of SiO_2 layers should be thicker than other organic layers to make perfect phase matching between optical pulses and THz waves [8, 13]. According to calculations at 6 THz in $OH1/SiO_2$ structure, if thickness of layers varies completely periodically, we can reach higher efficiency, i.e. thickness of both $OH1$ and SiO_2 is 100μm. This behavior is due to low absorption of THz

waves in $OH1$ crystal and good phase matching. Figures 2 and 3 show efficiencies of THz wave generation by DFG method at two frequencies 3 and 6 THz in multilayer structure and bulk crystals $OH1$, $DSTMS$ and $DAST$. Oscillations in the bulks show phase difference between THz and pump waves. On the contrary, in multilayer structures, plot of THz waves efficiency shows monotonous increase as a function of distance propagation. Horizontal portion of figure shows THz wave propagation in quartz layer [8]. Also there is no oscillation at 3 THz frequency in bulk $OH1$ crystal, due to perfect phase matching.

Table 5: Thicknesses of layers in studied structures of $DSTMS/SiO_2$ at 3 and 6 THz.

$DSTMS/SiO_2$(at $3THz$)

Layer number	Material	Thickness(μm)
1	$DSTMS$	49
2	SiO_2	73.5
3	$DSTMS$	45
4	SiO_2	67.5
5	$DSTMS$	43
6	SiO_2	64.5
7	$DSTMS$	42
8	SiO_2	63
9	$DSTMS$	41
10	SiO_2	61.5
11	$DSTMS$	40
12	SiO_2	60
13	$DSTMS$	38
14	SiO_2	57
15	$DSTMS$	36
16	SiO_2	54
17	$DSTMS$	30
18	SiO_2	45
19	$DSTMS$	30
20	SiO_2	45
21	$DSTMS$	30

$DSTMS/SiO_2$(at $6THz$).

Layer number	Material	Thickness(μm)
1	$DSTMS$	18
2	SiO_2	46.8
3	$DSTMS$	17.8
4	SiO_2	46.28
5	$DSTMS$	17.5
6	SiO_2	45.5
7	$DSTMS$	17
8	SiO_2	44.2
9	$DSTMS$	16.8
10	SiO_2	43.68
11	$DSTMS$	16.5
12	SiO_2	42.9
13	$DSTMS$	15.8
14	SiO_2	41.6
15	$DSTMS$	15.2
16	SiO_2	40.3
17	$DSTMS$	15.2
18	SiO_2	40.3
19	$DSTMS$	15
20	SiO_2	39
21	$DSTMS$	15

Figure 2 demonstrates that $DSTMS/SiO_2$ structure has higher efficiency with respect to $DAST/SiO_2$ structure because of low absorption of THz wave of $DSTMS$ and good phase matching of $DSTMS$ at 3 THz and large nonlinear susceptibility of $DSTMS$. Despite the fact that $OH1$ crystal has higher FM than $DSTMS$ and $DAST$, large absorption at 3 THz is an obstacle to increase the efficiency. Figure 3 shows superiority of $OH1/SiO_2$ structure with respect to two other structures at 6 THz frequency and hence shows that $DSTMS/SiO_2$ structure has larger efficiency than $DAST/SiO_2$ structure at this frequency. High susceptibility, good phase matching and very low absorption of $OH1$ crystal at 6 THz range with respect to $DSTMS$ and $DAST$ is responsible for these results.

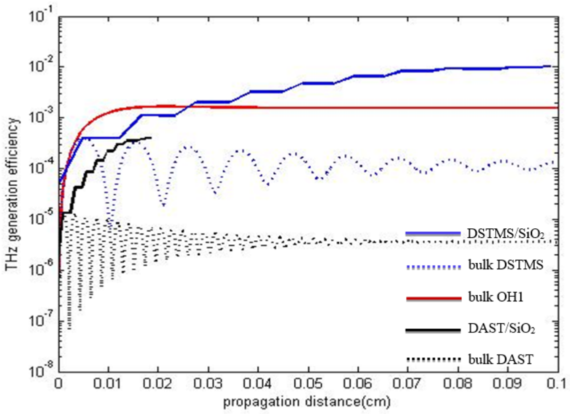

Figure 2: THz generation efficiency at 3 THz vs. propagation distance.

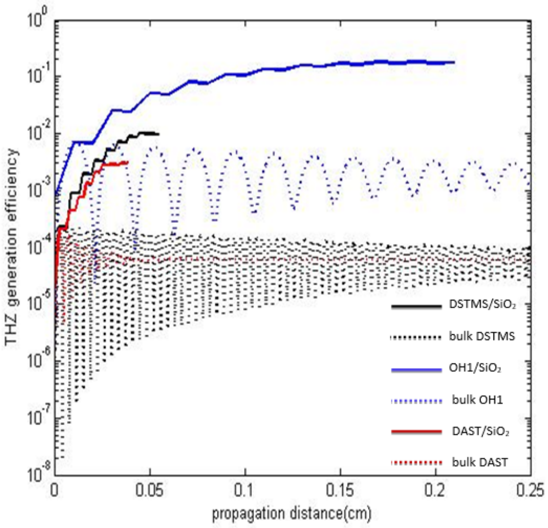

Figure 3: THz generation efficiency at 6 THz vs. propagation distance.

As Figures 2 and 3 show, efficiency in two structures, $OH1/SiO_2$ and $DAST/SiO_2$ at 6 THz frequency is higher than 3 THz frequency. Table 6 briefly demonstrates comparison of the above-described different multilayer structures efficiencies at two frequencies, 3 and 6 THz.

Table 6: Comparison of different multilayer structures efficiencies at two frequencies 3 and 6 THz.

Frequency (THz)	THz efficiencies of the proposed structures
3	$e_{DSTMS/SiO_2} > e_{OH1} > e_{DAST/SiO_2}$
6	$e_{OH1/SiO_2} > e_{DSTMS/SiO_2} > e_{DAST/SiO_2}$

It is noticeable that with increasing of input pump wave intensity we can increase output THz wave efficiency. Indeed, pump waves can be increased to the extent that they do not damage the crystal, so input wave amplitudes are valid in approximation for solution of differential equation (6). Figure 4 depicts output THz wave efficiency versus input pump wave. According to Figure 4, it can be concluded that there is a threshold intensity for input pump laser.

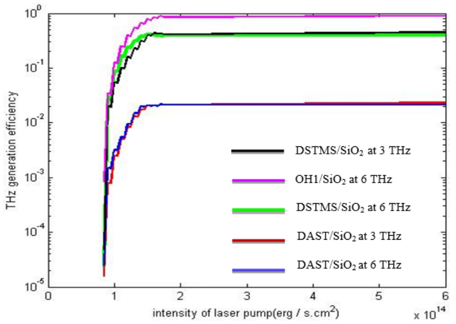

Figure 4: THz efficiency of output THz waves vs. intensity input pump laser.

It can be seen in three structures at two frequencies 3 and 6 THz, that up to $I = 2 \times 10^{14} erg/s.cm^2$, by increasing pump laser intensity, efficiency of generated THz waves increases, but after this value, increasing input pump wave intensity doesn't affect the output THz wave efficiency, and consequently it will be constant. $DSTMS/ZnTe$ structure is another structure proposed for enhancing THz wave efficiency and it causes efficiency enhancement of generated THz waves, but not monotonously. As seen in Figure 5 , this structure can not only enhance efficiency but also decrease efficiency because of high reflection of THz wave from the common surface at 3 THz.

In $DAST/GaP$ structure at 8 THz frequency, the reason is high reflection of THz waves from the common surface which makes $DAST/GaP$ structure inefficient and unsuitable to increase THz wave efficiency.

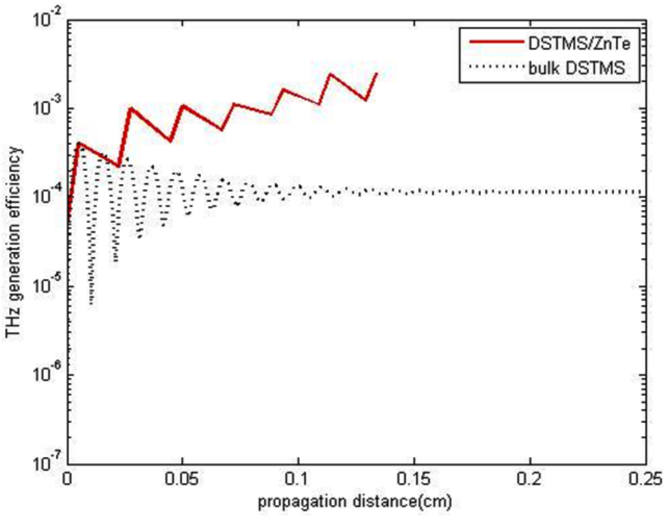

Figure 5: THz generation efficiency in $DSTMS/ZnTe$ structure at 3 THz vs. propagation distance.

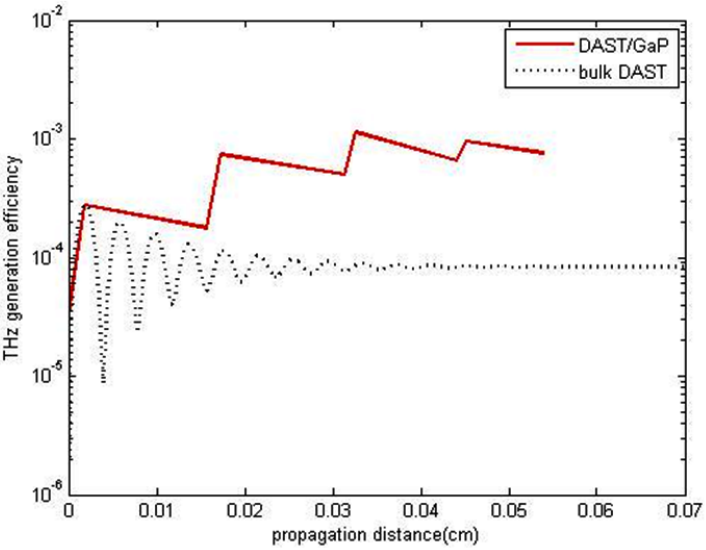

Figure 6: THz generation efficiency in $DAST/GaP$ structure at 8 THz vs. propagation distance.

3. Conclusion

Multilayer structures are more efficient than bulk crystals and we can increase efficiency of THz waves using them. Indeed, depending on the thickness of layers, absorption coefficients, phase matching and reflection from surfaces of all boundaries, one can obtain different efficiencies. Among all alternating structures, organic $crystal/SiO_2$ structures are more suitable, because of high electro optical coefficients of organic crystals. According to Table 6, at 3 THz frequency, efficiency of $DSTMS/SiO_2$ structure is higher than other multilayer structures and bulk $OH1$. On the other hand, at 6 THz frequency, efficiency of $OH1/SiO_2$ structure is higher than other structures. Meanwhile, two structures, $DSTMS/ZnTe$ at 3 THz and $DAST/GaP$ at 8 THz are not recommended as THz sources, due to their low efficiency and high reflection from interfaces.

Confilcts of Interest. The authors declare that they have no conflicts of interest.

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