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DAST/SiO_2 multilayer structure for efficient generation of 6 THz quasi-single-cycle electromagnetic pulses

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We propose a DAST/SiO_2 multilayer structure for efficient generation of near-single-cycle THz transients with average frequency around 6 THz via collinear optical rectification of 800 nm femtosecond laser pulses. The use of such a composite material allows compensation for the phase mismatch that accompanies THz generation in bulk DAST crystals. The presented calculations indicate a strong increase in the THz generation efficiency in the DAST/SiO_2 structure in comparison to the case of bulk DAST crystal.

OCIS codes: 190.7110, 300.6495.

During the past 10 years, significant progress has been made in the development of high-power THz sources [1]. Today, high-energy ultrashort THz pulses with average frequency below 2 THz can be obtained at large accelerator facilities [2] or through optical rectification of femtosecond laser pulses with tilted pulse fronts (TPF) using lithium niobate crystals as a nonlinear medium [3,4]. The TPF THz generation technique provides a laser-to-THz energy conversion efficiency of up to 0.25% [5,6], which corresponds to a photon conversion efficiency higher than 100%. In this way, THz pulse energies up to 125 μJ have recently been obtained using a table-top picosecond laser system [6]. Moreover, this technique allowed demonstrating a mobile source of 50 μJ THz pulses [7]. Lately, the efficient generation of high-power near-single-cycle pulses with average frequency around 2.2 THz through collinear optical rectification of 1.5 μm femtosecond laser pulses in an organic salt crystal, 4-N, N-dimethylamino-4′-N′-methyl-stilbazolium tosylate (DAST), has been achieved [8]. Tremendous progress has been made in the generation of broadband THz pulses using two-color pumping of a gas plasma. The laser-to-THz energy conversion efficiency has been increased from 10^{-8}, reported 12 years ago in the pioneering work of Cook and Hochstrasser [9], up to the recently achieved value of 10^{-4} [10]. However, this technique provides efficient generation at THz frequencies above 10 THz. High-power (up to 0.5 GW) few-cycle pulses with frequencies tunable from 10 to 72 THz were generated via difference-frequency mixing of two phase-locked femtosecond laser pulses [11]. Record power (15 TW) picosecond pulses at 28 THz were obtained by a CO_2 laser system [12]. In spite of this progress, there is still an absence of high-power table-top sources in the very attractive frequency range of 3–9 THz. 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 [13] and two-dimensional THz spectroscopy of the nonlinear vibrational response of water [14]. Thus far, high-power ultrashort THz pulses in this region have been exclusively provided by accelerator-based facilities [15].

Here, we propose the use of a DAST/SiO_2 multilayer structure for efficient generation of near-single-cycle 6 THz pulses. The use of quartz layers allows compensation for the phase mismatch that accompanies the generation of electromagnetic pulses with average frequency above 1 THz by collinear optical rectification of 800 nm femtosecond laser pulses in a DAST crystal oriented to provide maximum effective nonlinearity (d_{111}). Although the use of composite structures (ZnTe/GaP) for phase-matching improvement has been previously suggested [16], to the best of our knowledge, no model calculation or experimental demonstration of this approach has been provided so far.

DAST is a very attractive material for generating THz waves because of its high nonlinearity ($d_{111} =$ 615 pm V^{-1} at 800 nm) [17]. Around 1 THz, the effective nonlinearity of DAST is higher than that of $LiNbO_3$ and ZnTe by factors of 3.7 and 9, respectively [1]. However, because of the strong phase mismatch, collinear optical rectification of 800 nm pulses is not efficient in this material for the generation of THz pulses above 1 THz [18]. The TPF THz generation technique cannot be used in this material because, in contrast to $LiNbO_3$, the group velocity of laser pulses at 800 nm is lower than the phase velocity of THz waves. Employing 1.5 μm laser pulses instead of those at 800 nm provides an opportunity to substantially increase the generation efficiency of THz pulses with average frequency around 2.2 THz [8]. The use of an optical parametric amplifier for shifting the laser wavelength from 800 nm to 1.5 μm implies a decrease in the available pulse energy. Moreover, the effective nonlinear susceptibility of DAST at 1.5 μm is lower by a factor of 1.6 with respect to that at 800 nm [17].

An alternative approach to obtain high-efficiency THz generation from DAST is to engineer a multilayer structure for counteracting the phase mismatch appearing in different layers. In crystalline quartz, for example, the phase velocity of a THz wave over a broad frequency range is lower than the group velocity of 800 nm laser pulses [19,20]. Alternating layers of DAST and quartz of appropriate thicknesses (Fig. 1) can therefore avoid the phase mismatch that appears in a bulk DAST crystal.

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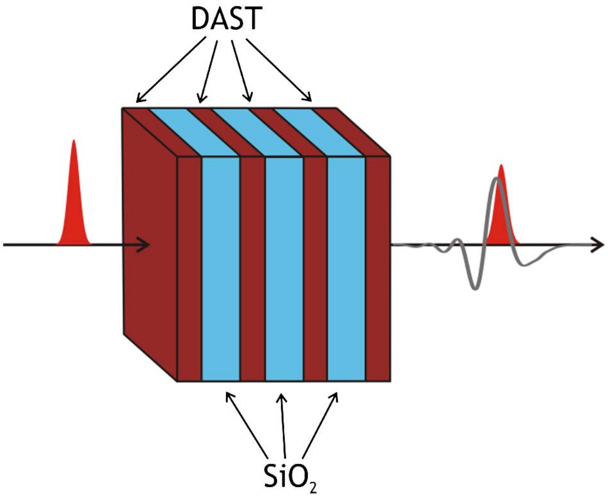

Fig. 1. (Color online) DAST/SiO$_2$ multilayer structure for efficient generation of few-cycle 6 THz pulses.

In the orientation providing the highest nonlinearity, the DAST crystal has a phase refractive index of 2.36 at 6 THz [21] and a group refractive index of 3.38 at 800 nm, whereas crystalline quartz at these frequencies has refractive indices of 2.2 and 1.55, respectively [20].

Using these figures, it is easy to estimate that the quartz layer should be thicker than the DAST layer by a factor of 1.6 in order to obtain full phase-mismatch compensation. Due to the relatively small difference in refractive index between DAST and quartz, the reflection of the generated 6 THz wave at the DAST/SiO$_2$ interface is only 0.1%. Moreover, the absorption of 6 THz radiation in crystalline quartz ($\alpha_q = 5$ cm^{-1} [20]) is small as compared to DAST ($\alpha_d = 140$ cm^{-1} [21]). The main factor limiting the number of layers in the structure is the reflection of laser light at the DAST/SiO$_2$ interfaces, which equals 4.5%. A structure consisting of 11 DAST and 10 SiO$_2$ layers would result in a decrease of the laser pulse energy by a factor of three (taking into account 16.5% reflection at the first DAST layer), which results in a ninefold decrease in THz generation efficiency since optical rectification is a second-order nonlinear process. In order to determine the optimal thicknesses for the DAST and SiO$_2$ layers and to estimate the THz generation efficiency, a model calculation has been performed. For the sake of simplicity, we consider a difference-frequency mixing of two monochromatic waves ($\lambda_1 = 793$ nm, $\lambda_2 = 806$ nm, $\Delta\nu = 6$ THz) in the undepleted pump approximation, taking into account the pump losses due to reflection at the DAST/SiO$_2$ interfaces and THz absorption in the DAST crystal. The nonlinear wave equation describing THz generation in this case is

$$\frac{dA_d}{dz} = -\frac{\alpha_d}{2}A_d - i\frac{2\omega_d}{n_d c} d_{\mathrm{eff}} A_{p1} A_{p2}^{*} e^{-i\Delta kz}, \qquad (1)$$

where A_d, A_{p1}, and A_{p2} are the complex amplitudes of the THz and laser pump fields, α_d and n_d are the THz absorption and refraction indices, ω_d is the THz angular frequency, d_{eff} is the coupling constant, and Δk is the wave vector mismatch ($\Delta k = k_1$-k_2-k_d). An analytical solution of Eq. (1) is

$$A_d = -i\frac{2\omega_d d_{\mathrm{eff}} A_{p1} A_{p2} e^{-i\Delta kL}}{n_d c\left[\frac{\alpha_d}{2} - i\Delta kL\right]} + A_i e^{-\frac{\alpha_d}{2}L}, \qquad (2)$$

where A_i is an integration constant used for tailoring solutions obtained for individual layers. Figure 2 presents the efficiencies of THz generation by difference-frequency mixing calculated for the DAST/SiO$_2$ multilayer structure (solid line) and bulk DAST (dotted line). The oscillations observed for the bulk DAST case are due to the phase mismatch between the THz and pump waves. Conversely, in the trace corresponding to the multilayer structure, one can observe a quasi-monotonous increase of THz generation efficiency as a function of propagation distance. In this case, the horizontal segments correspond to propagation of the THz wave in the quartz slices.

It should be noted that in our case (generation of 6 THz radiation by difference-frequency mixing of two waves with wavelengths near 800 nm), the generation efficiency of about 0.03 corresponds to the total depletion of one of the high frequency pump waves. For this reason, in the calculations, the typical experimental pump intensity (~100 GW/cm^2 [8]) has been reduced by a factor of 100 in order to maintain the validity of the undepleted pump approximation.

The calculations indicate that the highest THz generation efficiency is achieved by a smooth decrease in the thicknesses of the DAST/SiO$_2$ layers, going from 24/36 μm at the input face to 12/18 μm at the output face in the structure consisting of 11 DAST and 10 SiO$_2$ layers. This behavior results from the strong THz absorption in DAST, as shown in Fig. 3.

Around 6 THz, DAST does not have sharp absorption lines and its refraction index is very flat in this frequency range [21]. Similarly, the group velocity in DAST does not show strong frequency dependence around 800 nm [17]. For these reasons, the use of the proposed structure in conjunction with 800 nm laser pulses with pulse duration of about 50 fs instead of two CW waves would result in generation of near-single-cycle pulses at 6 THz with

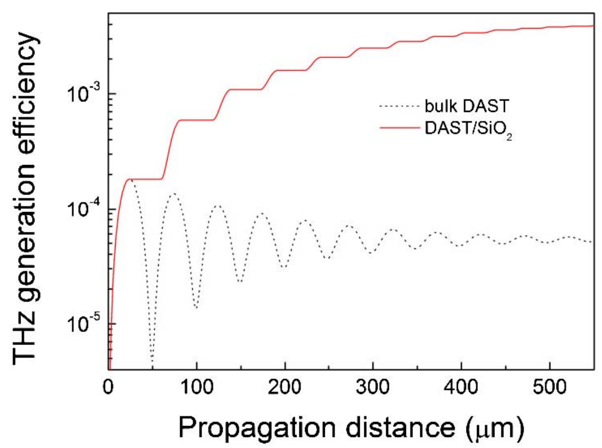

Fig. 2. (Color online) Generation efficiency of 6 THz radiation as a function of propagation distance of laser pump waves ($\lambda_1 = 793$ nm, $\lambda_2 = 806$ nm) in the DAST/SiO$_2$ multilayer structure (red solid line) and in bulk DAST (black dotted line).

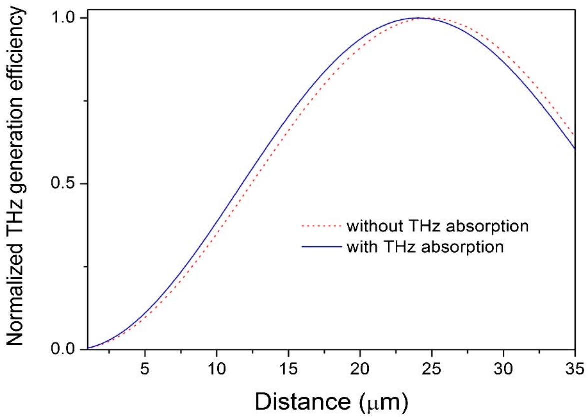

Fig. 3. (Color online) Normalized THz generation efficiencies in bulk DAST calculated both by including THz absorption (blue solid line) and neglecting it (red dotted line).

Fourier component parameters determine by the laser pulse duration and thicknesses of the DAST/SiO_2 layers.

Alternatively, the use of GaP instead of quartz in the structure would allow the central THz frequency to be increased to up to 8 THz. In fact, at this frequency, GaP has a phase refractive index of 3.68, whereas at 800 nm, it has a group refractive index of 3.55 [22]. One can estimate that, to obtain complete phase-mismatch compensation, the GaP layers should be thicker than the DAST layers by a factor of 8.8. In this case, we can expect an increase in conversion efficiency, because THz generation takes place in both layers. On the other hand, at 8 THz, DAST also has stronger THz absorption than at 6 THz [21]. It should be mentioned that the phase-matching technique discussed here substantially differs from the quasi-phase-matching technique based on using periodically poled materials [23,24], which was proposed in one of the pioneering publications of nonlinear optics [25]. In particular, the DAST/SiO_2 structure is aimed at generation of near-single-cycle THz pulses, whereas periodically poled materials provide an opportunity to obtain multicycle THz pulses.

In summary, we have proposed a new technique for the generation of high-power near-single-cycle THz pulses with a central frequency of 6 THz via optical rectification of 800 nm femtosecond laser pulses in a DAST/SiO_2 multilayer structure.

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