Research Article | Vol. 27, No. 22 / 28 October 2019 / Optics Express 32855

Optics EXPRESS

Double-pump technique – one step closer towards efficient liquid-based THz sources

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Abstract: By irradiating a water jet with double pulses, we demonstrate 4-fold higher THz wave generation than for a single pump pulse. The dependence of the enhanced THz signal on the temporal delay between two collinear pulses reveals the optimal time for launching signal pulse is near 2-4 ps, which corresponds to the time needed to create the complete pre-ionization state when sufficient electron density is already induced, and there is no plasma reflection of the pump pulse radiation. The increase in THz waves generation efficiency corresponds to the case of water jet excitation by the pulses with an optimal duration for a certain jet thickness, which is determined by the spatial pulse size. Using a theoretical model of the interaction of a high-intensity sub-picosecond pulse with an isotropic medium, we held a numerical simulation, which well describes the experimental results when using 3 ps value of population relaxation time. Thus, in this work, double pump method allows not only to increase the energy of the generated THz waves, but also to determine the characteristic excited state lifetime of liquid water. The optical-to-terahertz conversion efficiency in case of double pulse excitation of water column is of the order of $0.5{\cdot}10^{-3}$, which exceeds the typical values for THz waves generation during two-color filamentation in air and comparable with the achievable values due to the optical rectification in some crystals.

1. Introduction

Every year the interest in the terahertz (THz) frequency range is stimulated by the new ideas on its various applications. In addition to the common use in medicine, national security and imaging, radiation at these frequencies fits for wireless space systems [1,2]. Thus, the natural issue is to create economical and sufficiently powerful THz source for this purposes.

It was shown that laser filamentation-based methods have high potential for generating THz waves. Most of the works are devoted to the study of air filamentation [3,4], but it was demonstrated that the plasma channel induced in media with higher density (clusters [5], liquids [6]) can be the source of more powerful THz fields. This can be explained through more efficient ionization processes, as well as less critical [7] power. Water surrounds us everywhere - it is universal and financially economical, especially in view of high damage threshold in comparison with solid-state sources. The use of liquids as targets is promising since they provide renewed and smooth surface for each laser irradiation.

The phenomenon of the generation of THz radiation in liquid media has been rather fully considered in works [6,8]. The single-color excitation of acetone jet in the recent paper [9] gave an optical-to-terahertz conversion efficiency value comparable to the case of two-color laser filamentation in atmospheric air. Moreover, the authors of [10] have successfully modified the experimental setup replacing the flat liquid jets by liquid columns, thus, increasing THz generation

#377878 https://doi.org/10.1364/OE.27.032855
Journal © 2019 Received 13 Sep 2019; revised 17 Oct 2019; accepted 20 Oct 2019; published 25 Oct 2019

efficiency by an order of magnitude. However, such values are still insufficient for ubiquitous applications. This opens up a search for the new methods of enhancing the optical-to-terahertz conversion efficiency.

One of such methods can be double-pump medium excitation, which is often used for spectral analysis, X-ray generation and plasma studies [11–13]. In THz science, this technique has demonstrated its effectiveness for increasing the output power of THz radiation. In work [14], double pump method is used to increase the optical-to-terahertz conversion efficiency during laser filamentation in air. The physical basis of this approach is mainly that the signal beam interacts with weakly pre-ionized medium, thus enhancing ionization and increasing the ablated mass. The authors of [15], demonstrated the THz energy dependence on the focal position of the second pulse and 10 times higher optical-to-terahertz conversion efficiency values.

In this work, we experimentally and numerically investigate the double-pulse excitation technique to enhance the generation of THz radiation in case of laser single-color filamentation in water jet. In order to understand the character of the enhanced THz signal dependences on the temporal delay between two collinear pulses, we consider the cases of using different population relaxation time values in our numerical simulation. The theoretical analysis reveals the enhancement indeed depends on the pre-plasma effect, considering the excited state lifetime is on the picosecond timescale for a liquid water. Our results pave the way for further development of the high-power THz field generation technology important for both fundamental science and practical application.

2. Experimental setup

We use a similar experimental scheme presented in [6] with excitation laser wavelength of 800 nm, p-polarization, pulse energy up to 2 mJ, repetition rate of 1 kHz, and single pulse duration with values from 35 to 400 fs. The principal part of the set-up scheme is displayed in Fig. 1.

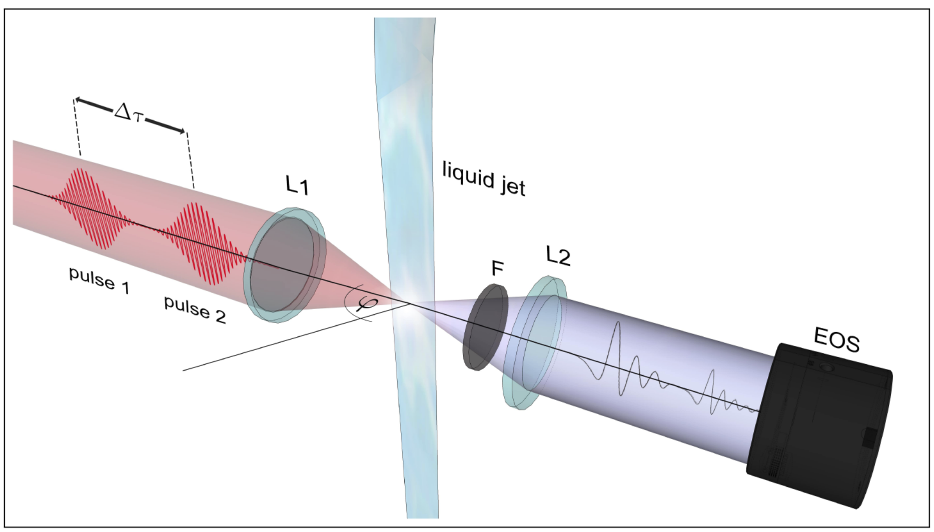

Fig. 1. The double-pump experimental scheme. The pump laser radiation (consisting of the reference and signal pulses with a temporal delay of $\Delta\tau$) is focused on the lens L1 with a focal length of 5 cm. The focused radiation further falls on the water jet at the optimal angle of incidence $\varphi = 60°$. The generated THz radiation, collected and collimated by TPX lens L2 and filtered by a teflon filter F, is registred on the standard electro-optical scheme EOS.

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The laser beam is divided by a beam splitter into the probe and pump parts. The latter is further split into two pump beams (reference and signal beam) of 0.45 mJ energy by a Michelson interferometer. After passing the interferometer, the pump is focusing with a 5 cm lens (L1) on the liquid jet. In this study, we use various jet configurations - flat water jets and water columns. In case of using flat liquid jet, we install it on a rotation stage, which allowed it to change the radiation angle of incidence. For liquid water the value of the optimal angle is $\varphi = 60°$ [8]. After filtering (F) and collimating (L2) the output radiation we register THz waves, using standard THz time-domain spectrometer (EOS) based on the electro-optical effect with 1 mm thick ZnTe crystal, which make it possible to detect a signal up to 3 THz. The flat liquid jet is obtained using the nozzle which combines the compressed-tube nozzle and two razor blades [16]. Due to the use of the pump, water is released under the pressure. The rate of flow (1 m/s) is sufficient for a complete water refreshment in the area of filament-medium interaction at 1 kHz laser repetition rate.

3. Double pulse excitation experimental results

3.1. THz radiation energy enhancement in flat water jets

To begin with, we study experimentally the enhancement of the THz radiation energy, generated during single-color laser filamentation, using double pulse excitation of a flat water jet (Fig. 2). At the constant energy for each of two pulses and variable pulse duration, THz radiation energy enhancement is measured relative to the temporal delay. In order to obtain the terahertz pulse energy, we integrate the square of the THz field amplitude in time.

The typical temporal forms of the THz signals, generated in the jet of water during double-pulse excitation are shown in Fig. 2(a). It explicitly demonstrates the enhancement of the signal pulse relative to the reference one. The corresponding THz spectra obtained using the Fourier transform are also included. Figure 2(b) demonstrates the THz waves energy dependence on the temporal delay $\Delta\tau$ between two pump pulses typical for the series of our experiments on THz generation in water jet of 100 and 270 μm thickness.

Figure 2(b) shows that the maximum enhancement value corresponds to the optimal duration for a certain jet thickness [9]. Moreover, using a thicker jet we get weaker THz signal due to the linear THz absorption in water medium. The general form of the THz enhancement dependences on the temporal delay between two collinear pulses is the same for all initial conditions. We get maximum enhancement value when the signal beam is launched approximately 2-4 ps after the reference one, which presumably corresponds to the time needed to create the complete pre-ionization state when sufficient electron density is already induced, and there is no plasma reflection of the pump pulse radiation [17]. Moreover, THz radiation energy enhancement is achieved with time delays up to 20 ps. Further attenuation indicates the influence of relaxation processes.

3.2. THz radiation energy enhancement in water columns

In the second part of the experimental study, we present enhanced THz signal measurements with double pulse excitation of water columns. The authors of work [10] introduced the experimental setup modification, replacing flat liquid jets by liquid columns. The benefit of using columns is in decreasing the total internal reflection of the THz wave at the liquid-air interface. The optical-to-terahertz conversion efficiency in these experiments reached the values of 10^{-4}. In this regard, we present measurements of the THz radiation energy enhancement during double pulse excitation in the water columns (Fig. 3). We use a jet of 210 μm thickness at the flow rate of 2 mL/min. The pump radiation is focused in the column center. The varied duration of the signal and reference pulses is equal to 61, 90 and 145 fs.

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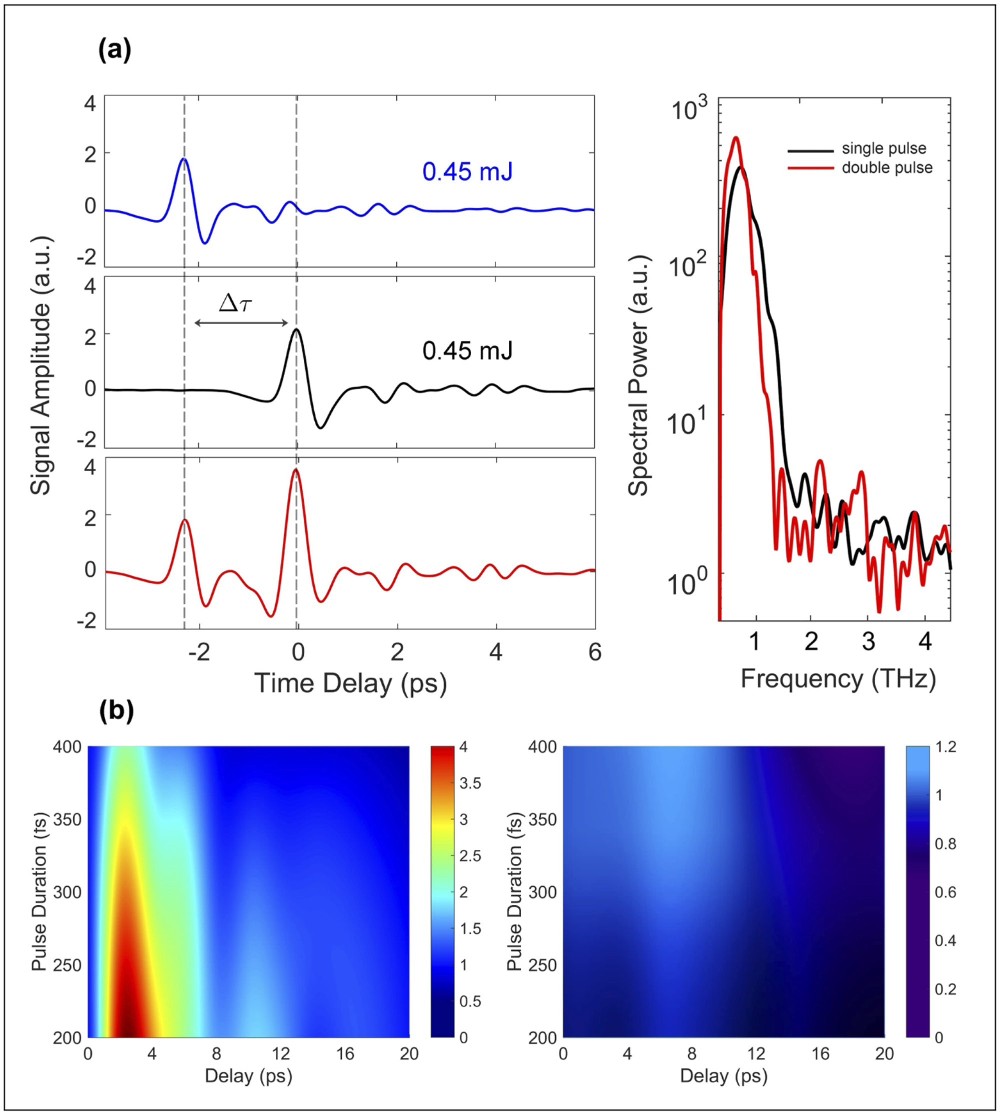

Fig. 2. a. The temporal forms of THz signals (left) generated during single-color double pulse excitation of a water jet with a temporal delay $\Delta\tau$ = 2.3 ps. The corresponding spectra are presented on the right. b. THz waves energy dependence on the temporal delay $\Delta\tau$ between two pump pulses during propagation in water jet with thickness of 100 μm (left) and 270 μm (right).

As it is in case of double pulse excitation in flat water jets, THz radiation energy is more than 4-fold higher comparing to single pulse excitation in water columns. Although modifying jet configuration, we get the same enhancement range up to 20 ps. An estimate of the conversion efficiency of $0.5{\cdot}10^{-3}$ is obtained by comparing the energy enhancement in case of single pulse [10] and double pulse excitation and exceeds the typical values for THz waves generation during two-color filamentation in air and comparable with the achievable values due to the optical rectification in some crystals. All of the above require some theoretical justification.

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4. Simulation results and discussion

In order to investigate the physics of THz waves energy enhancement under double pulse water jet excitation, we further consider the bulk nonlinearity. To numerically simulate the process under study, we use a theoretical model of the interaction of an ultrashort pulse with an isotropic medium [18]. We calculate the evolution of the optical pulse electric field using equations of the form:

$$\begin{cases} \frac{\partial E}{\partial z} - a\frac{\partial^3 E}{\partial \tau^3} + gE^2\frac{\partial E}{\partial \tau} + \frac{2\pi}{cn_0}j = 0 \\ \frac{\partial j}{\partial \tau} + \frac{j}{\tau_c} = \beta\rho E^3 \\ \frac{\partial \rho}{\partial \tau} + \frac{\rho}{\tau_p} = \alpha E^2 \end{cases} \tag{1}$$

where n_0 and a are introduced to describe the normal dispersion $n(\omega) = n_0 + ca\omega^2$ in liquids in range up to 1.2 μm; g characterizes low-inertia cubic nonlinearity and connected to medium nonlinear refractive index through $g = 2n_2/c$; ρ and j are excited states population of the medium molecules and a current density of quasi-free plasma electrons induced by a strong radiation field; $\alpha \cdot \beta$ characterizes the efficiency of electrons transition to quasi-free states; τ_c is collision relaxation time; τ_p is electron excited state relaxation lifetime and in the current research is defined in such a way that to fit the experimental results; z is the direction of propagation; $\tau = t - zn_0/c$ is time in the moving frame of reference , where t is time, c is the speed of light in vacuum.

All coefficient values are taken according to work [9]. In this work THz waves emission due to single pulse excitation of liquids was already discussed. The generation of THz radiation according to the presented theoretical model occurs due to spectrum superbroadening under the mutual influence of Kerr nonlinearity and self-induced plasma.

This equation should be supplemented by boundary conditions. To simulate the generation of THz radiation in the case of double-pump excitation, we assume that the normalized radiation field is yielded by two collinear chirped Gaussian pulses with the same energy and a variable temporal delay:

$$\tilde{E}(\tilde{\tau}) = exp(-(\frac{\tilde{\tau}}{\tilde{\tau}_{pulse}})^2)sin(\tilde{\tau} + A\tilde{\tau}^2) + exp(-(\frac{\tilde{\tau} - \Delta\tilde{\tau}}{\tilde{\tau}_{pulse}})^2)sin((\tilde{\tau} - \Delta\tilde{\tau}) + A(\tilde{\tau} - \Delta\tilde{\tau})^2) \tag{2}$$

where $\tilde{E} = E/E_0$, E_0 is a peak amplitude of the pulse at the input surface; $\tilde{\tau} = \tau < \omega >$, $< \omega >$ is central frequency of radiation; $\tilde{\tau}_{pulse} = \tau_{pulse} < \omega >$ is normalized pulse duration; A is a fitting parameter, representing a pulse chirp, that is chosen such that the width of the chirped pulse spectrum matches width of the spectrum for the 35 fs spectral-limited pulse, and $\Delta\tilde{\tau} = \Delta\tau < \omega >$ is a normalized temporal delay. The intensity value I = 10^{13} W/cm^2 is chosen on the basis of experimental data.

The numerical simulation of a modeled dynamic process is based on the predictor-corrector method for spatial steps calculation; the uniform discrete grid approximates time derivatives. For predictor step the second-order Nyström method is used; for correction – the second-order Henrici-Milne method. Numerical integration for integral terms is done by fourth-order Simpson formula [19].

Figure 4 shows the result of numerical simulation of THz waves energy enhancement using double pulse excitation method. We get two THz waveforms on the out while launching two collinear 200 fs pulses with energy of 450 μJ and 6 ps temporal delay (Fig. 4(a)).

The dependence of the signal pulse double-pump enhancement on delay $\Delta\tau$ for different characteristic relaxation time values τ_p is demonstrated in Fig. 4(b). The data here is normalized to the enhancement maximum.

As a result, the enhancement range increases with increasing the time of electron relaxation from highly excited levels. For τ_p=3 ps we get best fit to the experimental data (Fig. 4(c), dots). Thus, it follows the damping process for liquid water is on the picosecond timescale.

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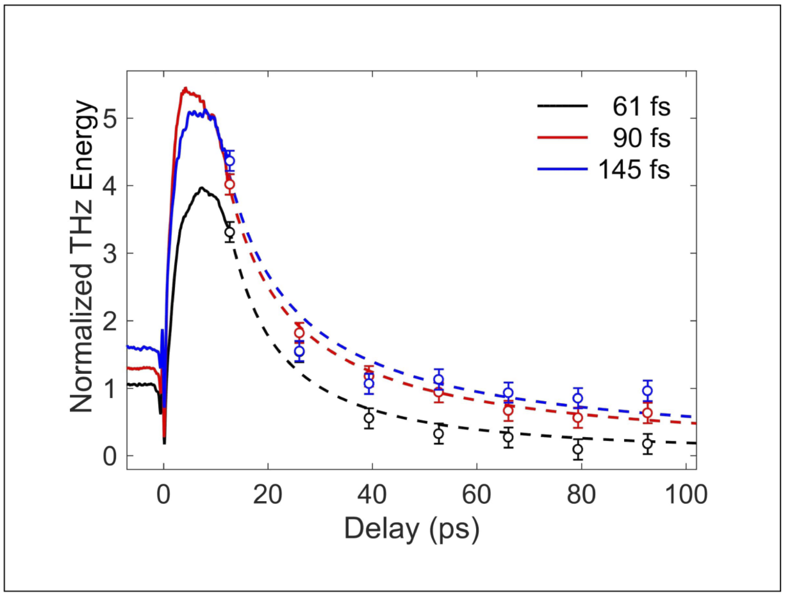

Fig. 3. THz waves energy enhancement dependence on temporal delay between two collinear pulses with durations of 61 fs (black), 90 fs (red) and 145 fs (blue). Dashed line corresponds to the exponential fit.

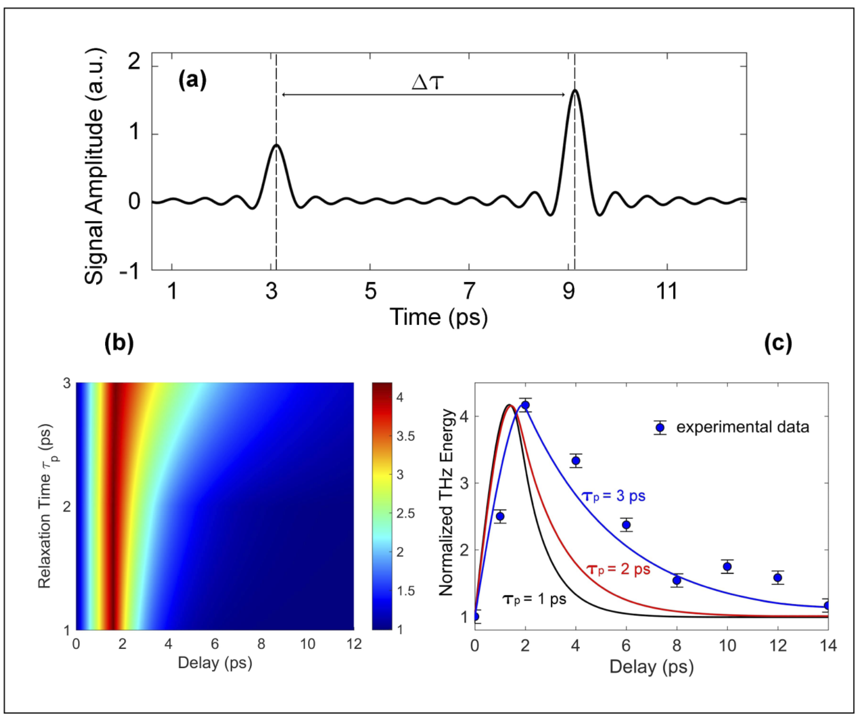

Fig. 4. a. Numerically simulated THz waveforms, generated while launching two collinear 200 fs pulses with energy of 450 μJ and 6 ps temporal delay. b. The dependence of the THz waves energy enhancement in the simulation of a single-color double pulse water jet excitation with the population relaxation time values from 1 to 3 ps. c. The correspondence of the experimental data (dots) to the result of numerical simulation for τ_p=3 ps.

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5. Conclusion

In conclusion, we observed the optical-to-terahertz conversion efficiency value up to 10^{-3} during single-color double pulse irradiation of water jet. The increase in THz waves generation efficiency corresponds to the case of water jet excitation by the pulses with an optimal duration for a certain jet thickness, which is determined by the spatial pulse size. Using a theoretical model of the interaction of the subpicosecond pulse high-intensity field with an isotropic medium, we held a numerical simulation which fits the experimental results and reveals the impact of the residual electron population on the enhancement effect. By irradiating water medium with two collinear pulses with variable temporal delay, we show the optimal time for launching signal pulse is 2-4 ps after the reference one. At this moment, there is no plasma reflection of the pump pulse radiation and the sufficient electron density is induced. Our study is a first step towards new ways for enhancing filamentation-based THz waves generation in liquids.

Funding

Russian Science Foundation (19-12-00097); National Science Foundation (ECCS-1916068); Army Research Office (W911NF-17-1-0428); Air Force Office of Scientific Research (FA9550-18-1-0357).

Disclosures

The authors declare no conflicts of interest.

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