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A Proposed Experimental Scheme for the Generation of Tunable THz Spectral at Narrow Bandwidth via Two Color Laser Induced-Plasma

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Brief Report

A Proposed Experimental Scheme for the Generation of Tunable THz Spectral at Narrow Bandwidth via Two Color Laser Induced-Plasma

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Abstract: Modifiable THz spectral shapes are important tools that facilitate the comprehensive study of phonon dynamics in condensed matter systems. The generation of narrow bandwidth THz spectra with tunable center frequency which are suitable THz forms needed to achieve such objectives are currently less studied from the table top laser-induced plasma emitters' perspective. This experimental research is aimed at developing a robust two-color laser induced plasma set-up comprising of a temporal pulse stretcher and an Optical Parametric Amplifier that generates chirped and wavelength tunable pulses respectively. By focusing and independently controlling the ω and 2ω arms of the chirped pulses resulting after the interaction with a β-BBO crystal, I aim to generate narrow bandwidth THz signal (from plasma) scalable at MV/cm intensity and tunable in a wide THz spectral range, in addition to varying the frequency ratio mix.

Keywords: narrow-bandwidth; two-color laser; tunable frequency

1. INTRODUCTION

THz radiation has opened up a lot of fascinating research in the area of condensed matter Physics following their discovery about two decades ago. Before the advent of THz radiation, light-matter interactions have been performed with optical radiation located in the Electromagnetic spectrum which are only able to probe superficial electronic-related properties of matters. However, deep lying low energy material phenomena which are beyond the reach of those optical radiation typically have frequency response within the THz regime, and as such THz radiation has since been used as an effective tool to study and characterize biomedical samples, superconductors, ferromagnetic and explosive substance, just to mention a few [1–3].

Although, a lot of research both at the large scale and table-top sources [4–6] have been dedicated to generate THz radiation with spectral frequencies extending up to several 10's of THz, only a relatively few studies have particularly being concerned about generating narrow bandwidth spectrum whose central frequencies can be tuned to a wide range. THz radiation of such properties are mostly relevant for fundamental research among others, owing to the fact that the THz spectral response of certain materials are found within narrow regions in the THz regime [7].

In order to maximize the applicability of the THz radiation sources, there is a need for a dynamic control over the characteristics of the radiated THz pulse, in that the spectral bandwidth of a broadband THz spectral can be narrowly adjusted, with the addition of detuning the center frequency over the THz spectral range. Other than the large scale sources of tunable narrow bandwidth THz radiation been generated at FLASH, FAST, THU [8–11] and among others, there are also few available table-top laser-based emitter sources such as PCA, nonlinear crystal and plasma operated at low cost [this work].

2. BACKGROUND STUDY

In the literature, the PCA's are among the foremost photonic based table-top sources for tunable narrowband THz radiation using laser sources. The output of the laser source are fed into a temporal pulse stretcher that modifies the pulse form. These resulting chirped pulses are split into two arms

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by a Mach-Zehnder interferometer (MZI), and the recombined signal having a quasi-sinusoidal pattern have been used to drive the PCA in order to generate 36 *GHz* bandwidth of THz spectra whose center frequency was tunable up to 1*THz*. The extent of the chirp and temporal delay between the chirped pulses are the key parameters that controls the bandwidth size and center frequency of the radiated THz pulses [12,13].

Broadly speaking, the irradiating laser source and the THz emitter are key parameters usually regulated in order to generate tunable narrow bandwidth THz signal of which stacks of periodically poled-$LiNbO_3$ (PPLN) and Quasi Phase Matched-GaAs (QPM-GaAs) crystals are among the prominently structured THz emitters used for this purpose. For such engineered materials, a simpler geometry of the pump pulses requiring no temporal stretching is utilized. In particular, the PPLN crystals have generated tunable 20 *GHz* THz radiation bandwidth, with a tunable 0.8-2.5 *THz* center frequency by controlling only the size and number of their domain without necessarily modifying the laser signal [14]. In addition to manipulating the THz center frequency, the possibility to achieve the same outcome was also realized by tuning the orientation of the PPLN to produce a variable 50-100 *GHz* bandwidth THz signal at center frequency ranging from 0.6-1.1 *THz* [15]. In the case of QPM-GaAs, where the length of the crystal and the wavelength of the pump pulses were observed to influence the bandwidth of the THz signal and its center frequency respectively, an efficient narrow bandwidth of 100 *GHz* centered between 1.1- 2.2 *THz* frequency were observed [16]. In a similar configuration involving the beating of the chirped pulses as in the case of PCA, a significant enhancement in the efficiency of the nonlinear crystal was recently demonstrated with the PPLN by independently controlling the relative spectral phase of the MZI output to generate THz signal at a center frequency of 0.361 *THz*, with 3.61 *GHz* bandwidth [17]. In the work of [18], a programmable pulse shaper has been used in place of the pulse stretcher to create multiple pulse train. The number of pulses and temporal spacing within the train respectively controlled the THz bandwidth and center frequency. In their work, they realized THz radiation in the frequency range of 0.5–2.0 *THz* at 200 *GHz* bandwidth with the use of ZnTe and GaAs crystals. Also, the beating of chirped pulses in a similar way as in the PCA with the addition of spatial modification to the laser pulse have been used to gate Lithium Niobate (NL) crystal in order to generate narrowband THz spectrum between 0.3–1.3 *THz*, and of bandwidth generally less than 0.1 *THz* [19].

Although, spectrally tunable THz signal have been observed, the strength of the signal are usually weaker compared to unshaped THz signal. The need for sufficiently high THz field strength and energy to stimulate nonlinear interactions for practical applications have only been realized with organic nonlinear crystals [20,21]. These crystals has the potential to yield intense THz pulses with wide spectra range. In the majority of the nonlinear-organic crystal related researches, the HMQ-TMS, DAST, DSTMS and OH1 crystals were popularly used. The pump pulses were either temporally modulated using zone plates or temporally stretched with grating pairs. In particular an intense THz peak power was observed in DSTMS and OH1 crystals with a characteristics bandwidth of 36-50 *GHz*, and a continuous tunable center frequency ranging from 0.5-7 *THz* [22]. Similarly, phase stabilized THz signal with energy as high as 1.9 *uJ* and a tunable frequency from 4-18 *THz* having a typical bandwidth less than 1 *THz* was reported in DSTMS [23]. In the work of [20], where DSTMS was also used, they reported a narrower bandwidth less than 0.5 THz however with a narrower spectral range between 0.5-6.5 *THz* and lower energy of 0.5 *uJ*. In another work, a different geometry was employed to split and recombine the chirped pulses using high and partial reflecting mirrors. This etalon system ensured a stabilized THz signal phase and as such THz frequency in the range of 0.3-0.8 *THz* with a maximum energy of 100 *nJ* was generated from HMQ-TMS crystal [21]. Following the results of [24], multicycle THz fields were radiated from DAST crystals when train of pulses generated from a zone plate were delivered to the crystal. By varying the thickness and tilt of the zone plate, THz frequency tunable from 2-5.3 *THz* were radiated from the DAST crystals.

Though tunable THz radiation have been observed from solid state materials (organic/inorganic nonlinear crystals and PCA), much less has been reported from laser-induced plasma. In the past, broadband high intense THz radiation have been observed from plasma induced by the ionization of two color laser pulses, from which *MV/cm* of THz field strength and *uJ* energy were measured. The

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two color laser induced plasma may therefore hold promising prospect towards nonlinear selective phonon interaction. At the present, there are no state of the art research in this area other than that reported by [25] where they have generated tunable THz radiation from a pulse shaper that supplied trains of square wave patterns to a BBO crystal. The bandwidth of the THz spectral increased from 1-3 *THz* as the THz central frequency was tuned from 2.5-7.5 *THz*. Though the size of the bandwidth is not an ideally narrow bandwidth for certain material inspection, it represents the foremost strive towards achieving spectrally tunable THz signal from laser-induced plasma table top sources.

In this work, we propose and plan to implement the concept of pulse chirping into the multiple color scheme popularly used for broadband THz pulse generation, where in particular, a flexible control of the individual colors can lead to the realization of narrow bandwidth THz spectral. By either controlling the frequency ratio or time lag between the colors, I aim to tune the center frequency of the THz spectra while maintaining the narrowness of the spectra bandwidth.

3. THEORY AND METHOD

The experimental set-up as shown in Figure 1.0 is divided into three main section namely; the pulse chirping, variable two- color mixing and Electro-Optic sampling (EO) detection section (not shown). The output of the 150 *fs*, 1 *KHz*, 2 *mJ*, 800 *nm* Ti:Sapphire laser is split by a beam splitter (BS) into pump and probe pulses, where, the probe will be used to carry out a scanning EO detection within a Gallium Phosphide (GaP) crystal. The pump pulse will be temporally stretched by two pairs of high-efficiency transmission gratings (1800 grooves/mm) so that the instantaneous laser frequency is chirped linearly in time across the pulse duration. The extent of chirping can be controlled by varying the separation distance between the grating pairs, while the insertion of the Spatial Light Modulator (SLM) in the Fourier plane is intended to suppress the presence of chirp effect in the generated THz signal. This temporally stretched pulse will be split by dichroic mirror DM_1, where one arm will be used as input for the Optical Parametric Amplifier (OPA) to generate several wavelengths, and the other arm used for incommensurate frequency mixing. The output of the OPA will be focused by lens L_1, onto a β-BBO crystal to generate its second harmonic.

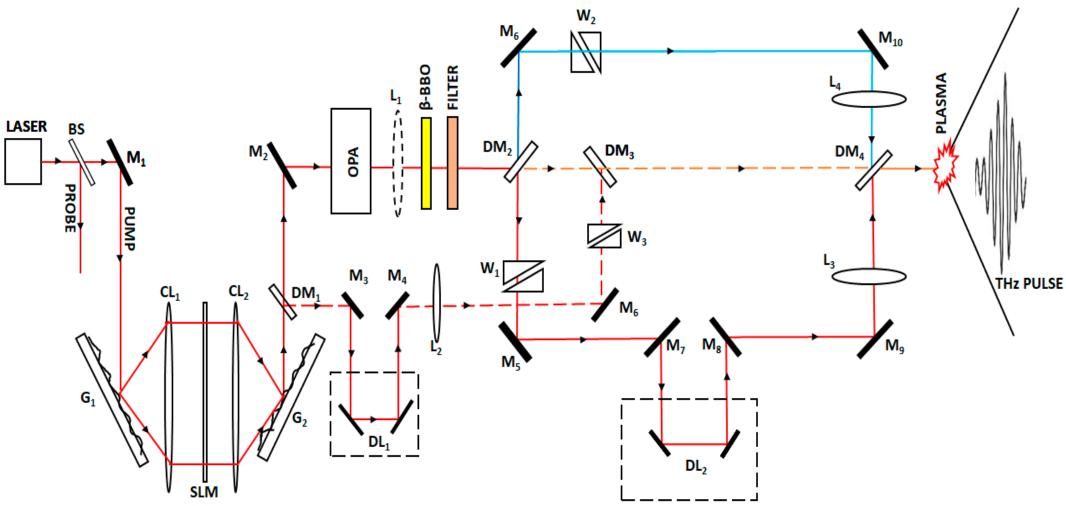

Figure 1.0. The sketch of the proposed experimental set-up for the generation of narrow bandwidth THz signal with the added flexibility of a widely tunable center frequency. BS: Beam Splitter; M_1, M_2, M_3, M_4, M_5, M_6, M_7, M_8, M_9, M_{10}: Mirrors; G_1, G_2: Grating pair; CL1, CL_2: Cylindrical lens; SLM: Spatial Light Modulator; DM_1, DM_2, DM_3, DM_4: Dichroic mirrors; L_1, L_2, L_3, L_4: lens; OPA: Optical Parametric Amplifier; β-BBO: beta-Borium Borate crystal; W_1, W_2, W_3: Wedge; Delay Line: DL_1, DL_2. *The figure is not drawn to scale.*

Another dichroic mirror DM_2, will split the ω and 2ω components into separated arms so that an independent control in their phase, intensity, polarization, and time lag can be realized for each component. Two wedges W_1 and W_2 are inserted to compensate for the Group Delay Dispersion and

Third Order Dispersion in each arm. The electric field for the two pulses with a relative delay τ, in one of the arm can be defined by,

$$E_1(t) = a.Re\left[e^{-\frac{t^2}{T^2}}e^{i\left(\omega_1 t + \frac{\alpha t^2}{2}\right)}\right], \text{ and } E_2(t-\tau) = b.Re\left[e^{-\frac{\sqrt{2}(t-\tau)^2}{T^2}}e^{i\left[\left(\omega_2(t-\tau) + \frac{\alpha(t-\tau)^2}{2}\right)+\phi\right]}\right] \quad (1)$$

Where ω_1 and ω_2 are the fundamental and harmonic frequency whose amplitude are defined by the term a and b respectively. T is the chirped pulse length which determines the THz bandwidth spectrum Δf_{THz}, according to $\Delta f_{THz} = \frac{\sqrt{8}\log(2)}{\pi T}$ **[26,27]**. Where T can be controlled by the extent of the chirp rate α, of the stretched pulse. The two beams are then recombined, and by neglecting their relative phase difference ϕ, the intensity cross correlation of the overlapping beams having a detuning frequency f_0, is given by;

$$I = Re\,|E_1 + E_2|^2 \sim \cos(\omega_2\tau - \alpha\tau^2 + f_0 t) \quad (2)$$

Where, $f_0 = \frac{|\omega_1 - \omega_2| - 2\alpha\tau}{2\pi}$ gives rise to a frequency modulated beam, and also serve as the deterministic factor for the center frequency of the emitted THz radiation. For a fixed chirp rate, it is straightforward to see that the center frequency of the generated THz signal can be controlled by either the temporal delay between the chirped pulses, or their difference frequency mixing. The set-up is designed to allow for commensurate and incommensurate mixing. In the case of commensurate mixing, DM_1 is flipped away so that the full power of the chirped pulse is sent to the OPA. Then the OPA is tuned to generate several wavelengths which will correspond to several *ω-2ω* mixing. In this configuration, the filter located next to the β-BBO crystal will be flipped away. While for the case of incommensurate mixing i.e., *ω-nω,* where *n* is a real number, the filter will be in place instead, whereas DM_2/ DM_4 will be flipped away. The output of the filter and the reflected beam from M_6 after been passing through W_3 are then combined in DM_3 and thereafter focused to generate plasma.

The ensuing plasma due to the beating of the chirped pulses is formed by the ionization of the surrounding air molecules resulting to the emission of fast moving electrons whose drift velocity V_d, is dependent on the form of the irradiating laser field as per [28];

$$I = Re\,|E_1 + E_2|^2 \sim \cos(\omega_2\tau - \alpha\tau^2 + f_0 t) \quad (3)$$

V_d, is the semi-classical velocity term that could also be derived by solving the Time-Dependent Schrödinger Equation, while *E* is the cross correlated electric field of the chirped pulses. The motion of the electron will give rise to a transverse electron current *J*, as per **[28]**,

$$J = \int e.V_d(t', t'').W_{ST}(t'').n_g\;dt'' \quad (4)$$

Where. W_{ST} is the static tunneling model for the ionization rate of the air molecules, n_g being the density of the gaseous medium. By applying the Photocurrent model, forward propagating and narrowband THz spectral E_{THz}, can be radiated according to **[28]**;

$$E_{THz} \sim \frac{\partial J}{\partial t} \quad (5)$$

In summary, it is important to note that, due to the inherent complexity of certain biological macromolecules such as amino acids, peptides, proteins, nucleic acids and carbohydrates, the absorption spectra of a broadband THz signal may not be able to effectively resolve their closely spaced vibrational modes, hence a broad absorption pattern usually without any obvious peaks are generally observed. By utilizing the narrowband THz spectra (that is expected to be generated), we envisage to perform spectroscopic studies to demonstrate its application in biomedical science among others.

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