Femtosecond Laser Driven Plasma for Terahertz Generation

B. Muller¹, F. Aljammal¹, <u>G. Gaborit^{1,2}</u>, M.Bernier¹, E. Herault¹, J.-L. Coutaz¹

¹Univ. Grenoble Alpes, Univ. Savoie Mont-Blanc, UMR CNRS 5130, Grenoble INP, IMEP-LAHC, Bât. Chablais,

73376 Le Bourget-du-Lac, France

²KAPTEOS, Alpespace, 73800, Sainte-Hélène du Lac, France

Abstract: In this paper we present a third order nonlinear process within photo generated plasma. By mixing two fundamental photons in the plasma with one associated second harmonic generation photon, we create one photon with energy in the terahertz band.

Keywords: Plasma photo generated, terahertz generation, optical rectification.

1.Introduction

The spectral range lying from microwaves to optical spectrum was known as the "terahertz (THz) gap" due to the lack of efficient sources and detectors in this spectral region up to the 90's. This had prevented any use of THz radiation particularly useful in spectroscopy since vibrational modes of molecules are involved in the THz range. THz spectroscopy provides therefore unique fingerprints of material of great interest (e.g. explosives, pharmaceuticals, drugs...) for security and medical applications. Since the emerging of femtosecond pulsed lasers, this gap has been filled by scientists and industrials. Time resolved spectroscopy setups historically used in THz range take advantage of either non-linear effects in electro-optic (EO) crystals or photoconductive effect in semiconductors to generate and detect THz using laser pulse [1-6]. Such techniques provide picosecond electromagnetic bursts presenting broadband spectrum spreading typically from 0.1 THz to few THz- but still limited. Indeed, photoconductive antennas based on semiconductors have limited bandwidths due to photocarrier lifetime, while EO crystals absorb a part of the generated THz leading to dips in the available THz spectrum. The use of diluted media like gases could therefore leads to larger bandwidth. The first solution was proposed by Dai et al. [7] who used plasma as a centrosymmetric nonlinear media for broadband THz generation and detection. This technique offers the possibility to get THz waves 1 to 2 orders of magnitude more powerful than classical methods. It also gives the possibility to get a very broadband spectrum, from 0.1 THz to several tens of THz. To generate such broadband THz radiation, a high-power femtosecond laser pulse is focalized in air until breakdown is reached: a plasma is generated. The electrons produced by this ionization process are accelerated by an intense optical field asymmetry. This asymmetry is enhanced by using part of the optical pulse whose frequency was previously doubled in a non-linear crystal. The same principle is used to detect the THz signal.

2. Terahertz generation in air

Terahertz generation from photo generated plasma is a rather complicated process, as it involves several physical phenomena, such as strong-field ionization and interaction coupled with plasma dynamics and nonlinear optic process. The two-colours laser-induced plasma can be understand as a nonlinear medium where a four-wave mixing occurs: 2 photons from the optical pulse at pulsation ω and 1 photon at pulsation 2ω are mixed to generate a photon at THz frequency. The expression of the THz electric field can be written as:

$$E_{THz} \propto \chi_{pla}^{(3)} E_{\omega}^* E_{\omega}^* E_{2\omega} \tag{1}$$

, where E_{THz} , E_{ω} and $E_{2\omega}$ are respectively the THz, the fundamental and the second harmonic (SH) electric field, and $\chi_{pla}^{(3)}$ is an effective third-order nonlinear susceptibility of the plasma. To generate the SH photons, we use a BBO crystal whose SH generation efficiency is linked to its second-order non linearity $\chi_{BRO}^{(2)}$:

$$E_{2\omega} \propto \chi_{BBO}^{(2)} E_{\omega}^* E_{\omega} \tag{2}$$

So,

$$E_{THz} \propto \chi_{pla}^{(3)} \chi_{BB0}^{(2)} E_{\omega}^{4} = \chi_{pla}^{(3)} \chi_{BB0}^{(2)} (P_{\omega} - P_{0})^{2}$$
(3)

, where P_{ω} is the mean power delivered by the amplified laser and P_0 is the threshold power above which the plasma is photogenerated.

3.Experimental setup and the THz pulse

The experimental setup used for the emission and the detection of THz pulse is schematised in Fig. 1. This setup is based on an amplified laser delivering an ultrashort optical pulse (pulse duration = 50fs) centred at 800nm every millisecond (1 kHz of repetition rate). The mean optical power delivered is 4 Watts leading to an energy of 4 mJ per pulse. By focusing such pulses, one gets optical E-field as strong as several tens of GV/m, which is much higher than the disruptive E-field value in air (3.5 Mv/m at atmospheric conditions): a photo generated plasma occurs (see picture in Fig. 1).

The THz generation is based on the two-colours breakdown process described in part 2, and the detection uses Zinc telluride (ZnTe) EO crystal. The EO detection scheme is classically used for years [8] and is based on the fact that the THz E-field induces birefringence modulation of the ZnTe crystal. If the optical probe and THz pulses are synchronized in the EO crystal, the polarisation state of the optical probe beam is modulated proportionally to the THz E-field. By simply using a polarising beam splitter, the polarisation state modulation is transduced in optical intensity modulation detected by a pair of balanced photodiodes. The intensity modulation rate, which is therefore directly proportional to the THz E-field, remains very low and is coherently detected thanks to a lock-in amplifier locked on the frequency of a mechanical chopper. By changing the time delay between the THz and optical probe pulses using a delay line, one rebuilds the THz pulse. Fig. 2 presents the THz waveform and its associated spectrum.



Fig. 1. Experimental setup based on two-colours air-breakdown terahertz generation. Off-axis parabolic mirrors (OAPM) are used for beam (optical and THz) shaping.



Fig. 2. Terahertz waveform generated by the experimental setup (schematised in Fig. 1), and coherently detected using electrooptic detection, and its associated intensity spectrum (inset).

4. Results and discussion

In this work, we focus on the THz peak, and we propose to study the evolution of its magnitude with the optical power of the pump beam to experimentally validate equation (3). We decline these experiments first in air and then by adding a gas flow of either nitrogen (N_2) or helium (*He*) perpendicularly to the optical pump at the focal point, in order to change the gas mixture from which the plasma is photo generated. Results are presented in Fig. 3, and clearly show that the THz generated carries information about the plasma properties (composition and density).



Fig. 3: THz E-field generated by two-colours air breakdown process in air (blue dots), in nitrogen (red dots), and in helium (green dots) versus the optical fluence at the focal point, and their associated quadratic fitting curves (dashed lines).

Quadratic fitting curves (dashed line in Fig. 3) are in good agreement with experimental measurements (dots in Fig. 3), validating experimentally the model given by equation (3). This is true up to a fluence of about 0.9 MW/cm^2 above which a saturation is observed regardless the gas mixture. This is partially explained by the defocusing effect induced by the plasma.

We can also see that the THz generation efficiency depends on the gas photo ionised by the laser pulses. The N_2 flow (red curve in Fig. 3) improves slightly the THz Efield in comparison to the reference curve (blue curve), which has been obtained without any added gas flow to the air. This is in accordance with the fact that the flow enriches in nitrogen a medium naturally made of 80% of N_2 , by replacing O_2 molecules whose ionisation energy is equivalent. In these both cases, plasmas are close in composition and in density, which is not the case when purging the air with a *He* flow. In this case we replace N_2 and O_2 molecules by a monoatomic gas whose ionisation energy is almost twice bigger. The addition of *He* flow clearly minimizes the amplitude of the THz generated (see green curve in Fig. 3).

5.Conclusion

The authors will present during the conference, short THz waveforms and their associated broadband spectrum generated and detected by air plasma techniques, and their dependence on the properties of the photogenerated plasmas. We would like to meet the plasma community in order to discuss our results and present the two-colours breakdown THz pulse generation as potential tool for plasma diagnosis.

6.References

[1] J. Morris, and Y. R. Shen, "Far-infrared generation by picosecond pulses in eletro-optical materials," Optics Communications, **3**, 2 (1971).

[2] D. H. Auston, K. P. Cheung, J. A. Valdmanis and D. A. Kleinmann, "Cherenkov Radiation from Femtosecond Optical Pulses in Electro-Optic Media," Physical Review Letters, **53**, 16 (1984).

[3] M. Van Exter, and D. R. Grischkowsky, "Characterization of an Optoelectronic Terahertz Beam System," IEEE Transactions on Microwave Theory and Techniques, **38**, 11 (1990).

[4] A. Fekecs, M. Bernier, D. Morris, M. Chicoine, F. Schiettekatte, P. Charetter and R. Arès, "Favrication of high resistivity cold-implanted InGaAsP photoconductors for efficient pulsed terahertz devices," Optical Materials Express, **1**, 7 (2011).

[5] M. Bernier, F. Garet, J.-L. Coutaz, H. Minamide, and Atsushi Sato, "Accurate Characterization of Resonant Samples in the Terahertz Regime Through a Technique Combining Time-Domain Spectroscopy and Kramers-Kronig Analysis," IEEE Transactions on Microwave Theory and Techniques, **6**, 3 (2017).

[6] F. Sanjuan, G. Gaborit, and J.-L. Coutaz, "Full electrooptic terahertz time-domain spectrometer for polarimetric studies," Applied Optics, **57**, 21 (2018).

[7] J. Dai, X. Xie, and X.C. Zhang, "Detection of Broadband Terahertz Waves with a Laser-Induced Plasma in Gases," Physical Review Letters, 97 (2006).

[8] G. Gallot, and D. Grischkowsky. "Electro-optic detection of terahertz radiation," Journal of the Optical Society of America B, **16**, 8 (1999).