

# Ultrabroadband spectroscopy for security applications

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**Abstract**—Ultrabroadband spectroscopy is a promising novel approach to overcome two major hurdles which have so far limited the application of THz spectroscopy for security applications: the increased bandwidth enables to record several characteristic spectroscopic features and the technique allows for remote detection. However, for real applications several parameters still have to be optimized. A comprehensive evaluation of the potential of this technique includes for example a detailed study of the generation process in an air plasma. We present some aspects of our joint theoretical and experimental evaluation of the technique for defense and civil security applications.

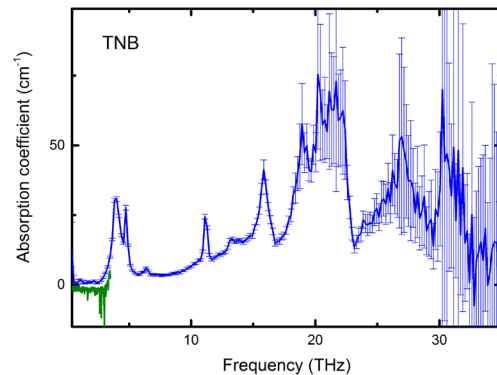
## I. INTRODUCTION

TERAHERTZ spectroscopy has proven to be a versatile tool for the detection of illicit substances like explosives [1], drugs [2] or counterfeit medicines [3]. Especially the detection of explosives has attracted much interest in the security and defense sector due to the major advantages THz technology has to offer, like penetration of non-conducting materials, minimal health-risk for operators and characteristic fingerprints of many security relevant materials at THz frequencies. However, typical THz systems offer just a limited bandwidth of up to 4 THz. For a non-ambiguous identification of dangerous materials, this limited bandwidth can be a serious problem, since, at least in explosives, many characteristic lines tend to show up at frequencies above 2 THz. Hence, in such systems just a small number of lines can be used for the identification of a material. However, since chirped-phase amplified laser systems nowadays deliver high intensities within femtosecond time scales, new mechanisms for creating THz radiation have become accessible, offering a strongly enhanced bandwidth [4]. Recent reports on two-color laser plasma THz generation and air biased coherent detection have attracted significant interest [5][6]. Such systems offer typically a bandwidth exceeding 40 THz, making it especially useful for the characterization and identification of different materials, including explosives [7]. First reports have thus sparked once again the interest in THz techniques for security applications. A detailed evaluation of the technique considering several parameters could thus help to confirm this potential.

## II. RESULTS

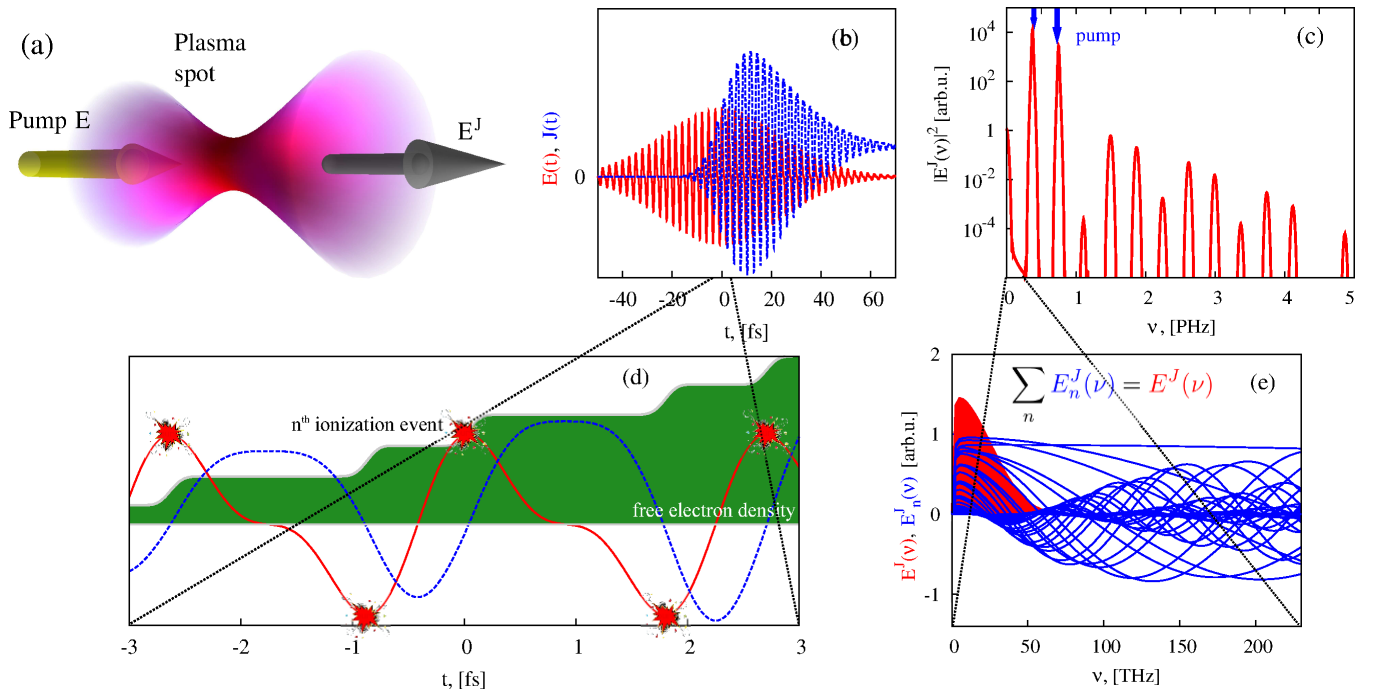
In a first step, we have updated our extensive data base of the spectra of a large variety of explosives, including new formulations of home-made explosives, by adding novel broadband spectroscopy data. Figure 1 shows the absorption coefficient of TNB as an example. The blue line gives the results obtained in the plasma based system, while the green

line is obtained in a conventional THz TDS system based on photoconductive antennas as emitter and detector. For better visibility a small offset has been introduced for the green line.



**Figure 1** Absorption coefficient of TNB (blue curves with error bars). For comparison the green line shows the result of a conventional THz TDS System.

The effects of several laser and gas parameters on the generation efficiency and spectral performances will be evaluated theoretically in view of further experimental validation. Preliminary results based on theoretical estimates and past numerical computations will be recalled. They mostly concern intense and broadband THz pulses generated by two-color laser pulses, e.g., a fundamental pulse and its second harmonic created in a frequency doubling crystal. We will discuss the possibility of increasing the energy yield in the spectral range between 1 and 100 THz for two optical pulse components co-propagating either in tightly focused geometry [8,9] or in filamentation regime in a noble gas and air [10]. Prior use of a noble gas (argon) is justified by the simplicity of the gas response compared to that of air components. Particular attention will be paid to pump pulses operating in the near infrared, i. e., at 800 nm. Our theoretical analysis emphasizes the importance of tunneling ionization [4,8,9], promoting a step-like increase in the electron density and asymmetric ionization instants favoring the creation of non-zero low-frequency charged currents responsible for broadband THz emission. Figure 2 gives a schematic overview on this two-color photocurrent mechanism. We will also discuss alternative potential mechanisms for THz generation such as longitudinal ponderomotive forces produced at high laser intensities [11]. We shall present semi-analytical evaluations of laser-driven THz spectra based on the so-called local current model in order to optimize and design the THz spectrum and fields through ionization-induced photocurrents.



**Figure 2** Mechanism and spectral properties of radiation generated by ionizing two-color pulses. (a) Schematic setup, (b) two-color pump field  $E(t)$  (red curve) consisting of fundamental and second harmonic frequency, and current  $J(t)$  (blue curve) induced by tunneling-ionization. (c) Spectrum of the generated radiation vs. frequency on a large frequency scale from THz to VUV. (d) Zoom into (b), showing the step-wise increase of the free electron density (filled green curve) by ionization events located at times  $t_n$  with extremal electric field  $E(t)$  (red curve). Free electrons produced in the various ionization events are then accelerated and produce the current  $J(t)$  (blue curve). (e) Zoom into the THz range of (c), explaining the spectral structure of the radiation  $E^J$  emitted by the current  $J$ . Each ionization event at time  $t_n$  induces broadband radiation  $E_n^J$  with comparable amplitudes but different phases (blue curves). The sum of all partial contributions yields the resulting spectrum  $E^J$  (filled red curve). The spectral structure of  $E^J$  is solely determined by spectral interference of different  $E_n^J$ .

The growth in the THz yield caused by an increase of the pump laser wavelength will be discussed [10]. The extreme sensitivity of THz radiation spectrum on the configuration of the femtosecond laser pulses can finally be exploited to tailor specific sources to the application. We numerically examine multi-colored pulse configurations extended to three and more optical frequencies, as these can be produced nowadays by optical parametric amplifiers. Since the relative phases of all frequencies and their respective energies can be in principle controlled, these degrees of freedom will be used to increase substantially the THz pulse energy by shaping the laser pulses in optimal waveforms, such as, e.g. sawtooth shapes [12].

### III. SUMMARY

In summary, we have measured the THz response of several explosives including TNT, TNB, RDX, HMX and ANTA, using a broadband plasma based THz spectroscopic system. The generation efficiency has been evaluated theoretically by varying laser-gas parameters in order to confirm the potential of the technique for increased performances for safety and security applications.

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