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Generation of widely tunable Fourier-transform-limited terahertz pulses using narrowband near-infrared laser radiation

Jinjun Liu, Hansjürg Schmutz, Frédéric Merkt *

Laboratorium für Physikalische Chemie, ETH-Zürich, CH-8093 Zürich, Switzerland

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ABSTRACT

Widely tunable, Fourier-transform-limited pulses of terahertz (THz) radiation have been generated using (i) crystals of the highly nonlinear organic salt 4-*N*,*N*-dimethylamino-4′-*N*′-methyl stilbazolium tosylate (DAST), (ii) zinc telluride (ZnTe) crystals, (iii) gallium phosphide (GaP) crystals, and (iv) low-temperature-grown gallium arsenide (LTG-GaAs) photomixers with THz spiral antennas. Outputs from two narrowband ($\Delta\nu < 1$ MHz, $\lambda \sim 800$ nm) cw titanium-doped sapphire (Ti:Sa) ring lasers with a well-controlled frequency difference were shaped into pulses using acousto-optic modulators (AOM), coupled into an optical fiber, pulse amplified in Nd:YAG-pumped Ti:Sa crystals and used as optical sources to pump the THz emitters. The THz radiation was detected over a broad frequency range and its bandwidth was determined to be ~10 MHz. The spectroscopic potential of the THz source is illustrated by the absorption spectrum of a pure rotational transition of OCS.

1. Introduction

Terahertz (THz) radiation, which lies in the frequency gap between microwave and infrared radiation and is loosely defined by the frequency range 10^{11}–10^{13} Hz, has recently found applications in imaging, spectroscopy, and material and biomedical analysis (see, for example, Refs. [1–5] and references therein). Although there have been significant advances in recent years, the use of THz radiation remains more limited than that of its neighboring bands in the electromagnetic spectrum, primarily because of difficulties associated with its generation and detection.

As a result of the limited availability of THz radiation sources, only little information is known to spectroscopists in this frequency range. Table-top sources of THz radiation are particularly attractive for use in experiments with supersonic beams and involving multiply resonant excitation schemes. Particularly desired would be narrowband tunable sources to study low-frequency vibrations of polyatomic molecules and molecular clusters, and the rotational motion of small molecules (see, for instance, Refs. [6–11] and references therein), and in the identification of possible molecular carriers of astrophysical spectra, for which there is considerable current interest in the context of the new observational facilities ALMA [12] and Herschel [13].

In recent years, several techniques have been developed to obtain spectroscopic data in the THz range. Whereas Fourier-transform spectroscopy is well suited to study the upper part of the THz range [14], backward-wave oscillator (BWO) based sources are attractive in the region up to 2 THz [9,15–18]. Quantum cascade lasers appear to be the most promising sources in the THz range but are still in the development stage [19–21]. The use of synchrotron radiation in combination with Fourier-transformation interferometers [22] and of free electron lasers [23] appears very promising but requires large-scale facilities. Broadly tunable coherent THz radiation can also be generated from visible or near-infrared (NIR) laser sources by difference-frequency generation (DFG) in nonlinear crystals and by photomixing in photoconductive materials, which represent the methods used in the present work.

The organic crystalline salt 4-*N*,*N*-dimethylamino-4′-*N*′-methyl stilbazolium tosylate (DAST) possesses very high nonlinear susceptibilities and electro-optic coefficients, which makes it one of the best nonlinear materials for THz generation and detection up to date [24]. THz radiation has so far been generated in DAST by optical rectification (OR) of a single fs laser pulse [25,26] and by DFG using two ns laser pulses [27–31]. By DFG of two ns laser pulses from a dual-wavelength optical parametric oscillator (OPO) with two potassium titanyl phosphate (KTP) crystals (1300–1800 nm, pulse energy 21 mJ, pulse duration ~10 ns, repetition rate 10 Hz), continuously tunable radiation over a wide frequency range (0.3–30 THz) with pulse energies up to 2.4 μJ (at 27 THz) has been demonstrated by Suizu et al. [29,30]. The bandwidth of the THz pulses so generated, however, was limited by the bandwidth of the OPO lasers (~70 GHz). Recently we reported DFG of widely tunable, pulsed, Fourier-transform-limited THz radiation (bandwidth ~10 MHz, peak power on the order of 100 μW) up to a frequency

* Corresponding author.
E-mail address: merkt@xuv.phys.chem.ethz.ch (F. Merkt).

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of 11 THz limited only by the detection method from pulse-amplified NIR ($\lambda \sim 800$ nm) laser radiation using a DAST crystal that was cut to fulfill the phase-matching condition [31]. In the present paper, we report the experimental details of the THz source and, for comparison, the generation of THz radiation using an as-grown (*c*-cut) DAST crystal.

Other nonlinear optical crystals than DAST can be used as THz emitters, and many such crystals have already been tested experimentally. One important category of THz crystals consists of semiconductors, such as zinc telluride (ZnTe) and gallium phosphide (GaP). Ref. [32] gives a comprehensive theoretical analysis of THz generation by DFG in semiconductor crystals.

Photoconductive antennas represent a further important class of THz emitters. First used by Brown et al. in 1995 to generate radiation up to 5 THz [33,34], low-temperature-grown gallium arsenide (LTG-GaAs) has been so far the most frequently used of all photoconductive materials. Mouret et al. reported narrow-bandwidth radiation up to 3 THz generated by mixing two cw Ti:Sa laser beams using a LTG-GaAs photomixer vertically integrated in the middle of THz spiral antennas [35–37].

Following our previous paper [31], we describe here a technique to generate widely tunable Fourier-transform-limited nanosecond THz pulses using several THz emitters, including nonlinear crystals of DAST, ZnTe and GaP, and LTG-GaAs photomixers. The laser-based source relies on the use of Fourier-transform-limited NIR pulses of programmable shape and length [38–40]. Its narrow bandwidth, high absolute frequency accuracy, and tunability through the entire THz range make it a promising radiation source for THz spectroscopy. The emphasis of this article is placed on the characterization of these properties and on a comparison of the THz emitters listed above.

2. Experimental setup

The experimental setup used for the generation of THz radiation and its detection is displayed schematically in Fig. 1. THz radiation was produced by DFG of two amplified narrow-bandwidth laser pulses in nonlinear optical crystals and by photomixing of the laser pulses using photoconductive antennas. The laser system, consisting of two Fourier-transform-limited pulse-amplified NIR lasers, has been described in previous publications [38–40] and only slight modifications have been introduced for the present investigation. Two Ti:Sa ring lasers (Coherent, 899-29 and 899-21, output power $300 \sim 400$ mW), both pumped by diode-pumped, frequency-doubled Nd:YVO_4 lasers (Coherent, Verdi, 532 nm, output power 5 W) and both operated near 800 nm, were used to provide single-mode narrow-bandwidth ($\Delta\nu < 1$ MHz) cw radiation. Through combinations of a Fresnel rhomb and a polarization beamsplitter, the outputs from both ring lasers were split into two components, one of which was used for frequency calibration and/or frequency stabilization, the other for THz generation. The frequency of the first laser (899-29) was tuned and calibrated, while that of the second laser (899-21) remained locked. The Doppler-free saturation absorption spectrum of molecular iodine in a 75-cm-long cell heated to 900 K [41,42] was used for the calibration of the absolute frequency of the first laser (899-29), while a confocal Fabry-Pérot (FP) étalon (free spectral range, FSR = 161.653(3) MHz and finesse = 10) was used for the relative frequency calibration. The frequencies of the hyperfine components of the I_2 transitions were determined with an accuracy of better than 100 kHz by comparing with positions measured using a frequency comb [42] in a separate measurement. The FSR of the étalon was determined using two such I_2 lines. The frequency stabilization of the second, fixed-frequency laser (899-21) was achieved by dithering the laser frequency around, and locking it to, the transmission maximum of another FP étalon (FSR = 149.966(3) MHz, also determined using the I_2 lines, and finesse = 30) by adjusting the driving voltage controlling the position of a Brewster plate placed inside the laser cavity using a home-built proportional-integral-derivative (PID) controller. The FP étalon itself was locked to a polarization-stabilized He–Ne laser (SIOS Messtechnik GmbH, SL-02/1). The output from the He–Ne laser was dithered by an acousto-optic modulator (AOM, Brimrose, GPF-1000-500-800), sent through the FP étalon and monitored by a photodiode. A correction signal was then sent to a piezoelectric transducer (PZT) mounted on one of the two mirrors of the FP étalon to lock the étalon to the transmission maximum of the He–Ne laser using another PID controller. The frequency stability of the 899-21 laser was estimated as the half width at half maximum (HWHM) of the FP étalon fringes (~2.5 MHz) over one day. The absolute frequency of the lock position was measured by a wavemeter (Burleigh, WA-1500) with an accuracy of 75 MHz.

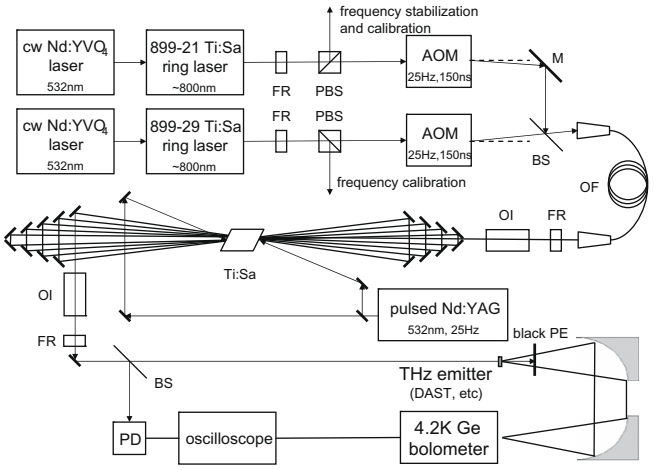

Fig. 1. Experimental setup for the generation and detection of THz radiation. FR, Fresnel rhomb; PBS, polarization beamsplitter; AOM, acousto-optic modulator; M, mirror; BS, beamsplitter; OF, optical fiber; OI, optical isolator; PE, polyethylene; PD, photodiode. Only one Ti:Sa amplifier is shown in the graph although two were used whenever pulse energies of more than 1 mJ were needed.

The other component of the cw radiation from each of the two ring lasers was sent to an AOM where pulses of adjustable duration and programmable shape were generated at a repetition rate of 25 Hz using an arbitrary waveform generator (Agilent, AWG-33250A). For the present experiment, the pulse shape was chosen to be a square function of 150 ns duration. The first-order sidebands of both laser beams, coming out of the AOMs with a 1 GHz frequency shift, were combined using a 50% beamsplitter and coupled into a single-mode polarization-maintaining optical fiber, which improved the mode matching and spatial overlap between the two lasers required for optimal THz generation. The temporal overlap of the two lasers was achieved using delay generators (Stanford Research DG535). The radiation pulses exiting the optical fiber had a peak power of ~20 mW. These pulses were guided into a nine-pass Ti:Sa amplifier consisting of a Ti:Sa crystal, pumped at a repetition rate of 25 Hz by the second harmonic output of a Nd:YAG laser (Quanta Ray, Pro270, 532 nm, pulse energy 200 mJ, pulse duration 10 ns). Each pass gave an amplification factor of about 3. Another six-pass amplifier, pumped by the same Nd:YAG laser (pulse energy 200 mJ), was used when higher pulse energies were needed. The maximum pulse energy of the final output was ~40 mJ, i.e., 20 mJ for each NIR frequency component. A Fresnel rhomb and an optical isolator were inserted between the optical fiber and the first amplifier as well as after the two amplifiers to prevent back reflections and to suppress amplified spontaneous emission (ASE). Similar techniques to generate narrow-band