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Intensity and phase retrieval of IR laser pulse by THz-based measurement and THz waveform modulation

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ABSTRACT

THz radiation is of great interest for a variety of applications. Simultaneously with the demonstration of high-intensity THz sources the idea to use this radiation for particle acceleration started to be investigated. THz accelerating gradients up to GV/m have been demonstrated in laboratory. THz radiation can be generated through the optical rectification process induced in non-linear crystals by a pump laser. The temporal shape of the pump laser and in general its characteristics are important aspects to be known in order to produce THz radiation via optical rectification in a controlled way. Here we present a technique that can be used to retrieve the temporal profile characteristics (envelope and phase) of the pump laser, starting from the detection of the THz waveform/spectrum and the knowledge of the physical/optical properties of the crystal used to produce it. This work also shows that the THz field can be shaped by properly acting on the pump laser phase. The possibility to opportunely shape the THz field is of great importance for many applications. Therefore this work paves the way to the possibility to coherently and dynamically control the THz field shape.

1. Introduction

The THz radiation is becoming of common use for many different applications, in particular the THz produced by non-linear crystals, organic and non-organic ones. Among the different processes for THz generation [1] the Optical Rectification (OR) is largely adopted in many laboratories. By the OR process is possible to convert an infrared laser pulse into a single-cycle THz one that can be useful for different applications: quantum control of materials [2–4], plasmonics [5–8], tunable optical devices based on Dirac-electron systems [9], medical imaging, security [3,10] and this started to be in use also in accelerator physics [11–14]. One of this novel application is the THz-driven linear accelerator as suggested in Ref. [12], another one is the THz-driven electron gun as in Refs. [11,13,14]; in both those new techniques the authors exploit the high electric fields of the THz for the acceleration of electrons. For most of those applications it is important to know the properties of the emitted THz radiation i.e. central wavelength of the spectrum, the electric field shape and the phase. These THz characteristics depend on the optical properties of the crystal, on its length and on the characteristics of the pump laser i.e. spectral amplitude and phase. Therefore the knowledge of the pump laser spectral phase and intensity is important to control the THz properties.

In this work it is demonstrated that it is possible to reconstruct the intensity profile and the spectral phase of the pump laser by measurements of the THz electric field together with the spectral amplitude of the pump. Moreover it is shown how the pump spectral phase affects the THz waveform resulting in a shaping of the THz electric field.

2. THz generation by optical rectification

The description of the OR process is done by writing the Maxwell wave equation for the electric field of the THz radiation in the case of a laser pump with a linear polarization:

$$\nabla^2 E_{THz} - \frac{\varepsilon}{c^2}\frac{\partial^2 E_{THz}}{\partial t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2 P_{OR}}{\partial t^2} \quad (1)$$

where ε is the complex first order relative dielectric function, that takes into account both the propagation and the absorption of the E_{THz} field, and P_{OR} is the non-linear polarization term which describes the Optical

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Rectification [15]:

$$P_{OR}(z,t) = 4\varepsilon_0 d E_p\left(t-\frac{z}{v_g}\right)E_p^*\left(t-\frac{z}{v_g}\right)e^{-\alpha z} \quad (2)$$

By Fourier Transforming the Eq. (1) with respect to the time variable and expliciting the OR term:

$$\nabla^2 E_{THz}(z,\omega) - \frac{\omega^2\varepsilon(\omega)}{c^2}E_{THz}(z,\omega) = = -\frac{4d\omega^2}{c^2}\left(E_p \star E_p^*\right)(\omega)e^{i\frac{\omega z}{v_g}}e^{-\alpha z} \quad (3)$$

where the symbol "⋆" is for the convolution in the frequency domain, v_g is the group velocity of the pump, α the absorption coefficient at the pump frequency, d is the effective non-linear susceptivity of the non-linear crystal, E_p takes into account for the envelope and the carrier frequency of the pump. The complex quantity $\omega^2\varepsilon(\omega)/c^2$ is redefined as $k^2(\omega)$ in the following.

Using the plane wave approximation for the forward propagating part of the Eq. (3) and imposing $E_{THz}(0,\omega) = 0$ for the complete solution we obtain:

$$E_{THz}(z,\omega) = TF(z,\omega)\cdot\left(E_p \star E_p^*\right)(\omega) \quad (4)$$

where $TF(z,\omega)$ is noted as the Transfer Function of the crystal in use, depending only on the characteristics of the crystal and its length L and it is defined by:

$$TF(z,\omega) = \frac{2d\omega^2 e^{ik(\omega)z}}{k(\omega)c^2}\frac{\left(e^{i(\frac{\omega}{v_g}+i\alpha-k(\omega))z}-1\right)}{\frac{\omega}{v_g}+i\alpha-k(\omega)} \quad (5)$$

In the OR process the crystal behaves like a frequency filter through the TF that depends on the thickness of the crystal and on its optical response at the frequencies of interest (IR and THz). In particular, the filtering effect depends on the length L of crystal in such a way that the longer the crystal is, the smaller is the bandwidth of the convolution product that will be converted into the THz domain. Since the bandwidth of the convolution product depends on the pump laser bandwidth, it is possible to find a maximum bandwidth of the pump that can be converted into the THz domain for a given crystal i.e. a given TF. A longer bandwidth will be affected by a loss of information due to the filtering effect.

It is possible to write the convolution in the frequencies domain as:

$$\left(E_p \star E_p^*\right)(\omega) = \frac{2I_p(\omega)}{\varepsilon_0 n(\omega_0)c} \quad (6)$$

where $n(\omega_0)$ is the index of refraction and $I_p(\omega)$ is the Fourier transform of $I_p(t)$.

Every crystal has a different frequency response resulting in a different conversion efficiency for every different pump central wavelength. Organic crystals (DAST, DSTMS, OH1) are well matched, from the conversion efficiency point of view, by laser pulses with central wavelength at 1225 nm. Laser pulse with central wavelength at 800 nm works better for non-organic crystals as for example ZnTe. In Fig. 1 some TF for different crystals of length of 0.5 mm are shown.

3. Intensity reconstruction procedure

The THz measurements shown in Fig. 2 are described in [16,17]. The pump laser used for the experiment was a Cr:forsterite (Cr:Mg_2 Si O_4) with a duration of τ = 95 fs Full Width Half Maximum (FWHM), measured through a standard autocorrelation technique, and a spectral bandwidth $\Delta\lambda = 27$ nm at around $\lambda_0 = 1225$ nm Fig. 2. This corresponds to an almost gaussian transform limit pulse with a time-bandwidth product $\tau\Delta\nu = c\tau\Delta\lambda/\lambda_0^2 \sim 0.49$, where for a perfect gaussian pulse the product value is ~ 0.44. Therefore the pump was neither chirped nor affected by higher order dispersion modulations. The lengths of the different crystals used during the OR experiment, in Ref. [16], were:

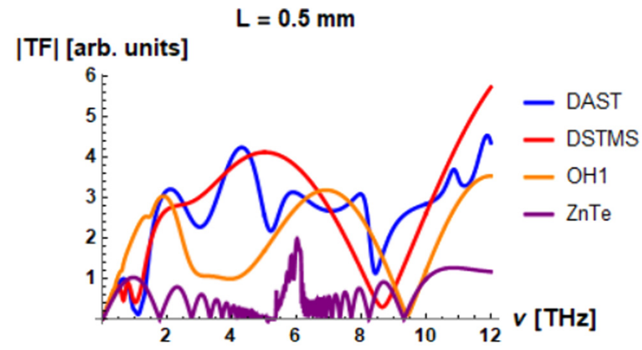

Fig. 1. Modulus of the transfer function $TF(L,\omega)$ (in arbitrary units) for the crystals ZnTe, DAST, DSTMS and OH1 in the range 0 – 12 THz. The crystals are considered to have the same length $L = 0.5$ mm.

0.18 mm for the DAST, 0.9 mm for the DSTMS and 0.44 mm for the OH1.

If we exploit the experimental THz spectrum shown in Fig. 2 and the transfer function of the 3 different crystals, it is possible to reconstruct the laser pump intensity by inverting Eq. (4) together with (6). The results of the reconstruction are shown in Fig. 3, where the 3 reconstructed intensity profile for the 3 different crystals and the input pump intensity taken from [17] are shown.

It is necessary to highlight that, in order to get clean and reliable results, as those shown in Fig. 3, we had to take into account the THz absorption from the water in air, (also called atmospheric absorption [18]) and for the losses due to the finite acceptance/chromatic effects associated to mirrors and lenses on the THz beam path from the source to the diagnostics. To do this, we retrieved the effect of the atmosphere and of the optical components with an algorithm that starting from the autocorrelation measurement of the pump reported in Fig. 2, calculated the expected THz spectra for the different crystals. Cross-comparing the expected spectra with the measured ones of Fig. 2 enabled us to reconstruct a correction to the transfer function that takes into account any loss due to the experimental setup. (see Fig. 4). The effective transfer function used in the retrieval is therefore:

$$TF_{eff} = TF_{atm}(z_{atm},\omega)\times TF_{op}(\omega)\times TF(z,\omega) \quad (7)$$

where $TF_{atm}(z_{atm},\omega)$ is the transfer function associated to a propagation length in the atmosphere (z_{atm}) and $TF_{op}(\omega)$ is the transfer function associated to the optics on the beam path, including specifically mirrors and lenses. In principle, given the atmospheric absorption index at fixed humidity α_{atm}, the function TF_{atm} can be determined as $\exp\left(-\alpha_{atm}z_{atm}\right)$. For what concerns instead the determination of the function TF_{op} and the THz losses due to the diffraction, a wave optics approach is generally needed [19], and the effect of each optic of the set-up can be taken under consideration from its transfer function provided by the manufacturer (and that depends principally on the coating quality, the geometry and size of the component). Therefore, in general, based on a specific optical setup, the effective transfer function can be known a priori.

The results shown in Fig. 3, are in good agreement with the reference intensity profile; for DAST and DSTMS crystals, the retrieved FWHM was 95 ± 15 fs in accordance with the autocorrelation measurement 2. In the OH1 case the reconstructed intensity is larger than the real one, with a FWHM of 126 ± 21 fs. This can be explained by a too long thickness of the OH1 crystal that makes the TF cut the bandwidth of the pump convolution at around 3 THz. Since the tails of $I_p(\omega)$ extends significantly up to about 7 THz, this cut causes a loss of information. The consequence is a greater value of the retrieved temporal FWHM. The relative error on the reconstruction technique is dominated by the noise fluctuations on the detected THz signals.

It is worth stressing that Eq. (4) holds for any kind of pump pulse $I_p(\omega)$, with any kind of spectral phase as shown in [20]. A direct consequence is that the THz field emitted via OR preserves the information

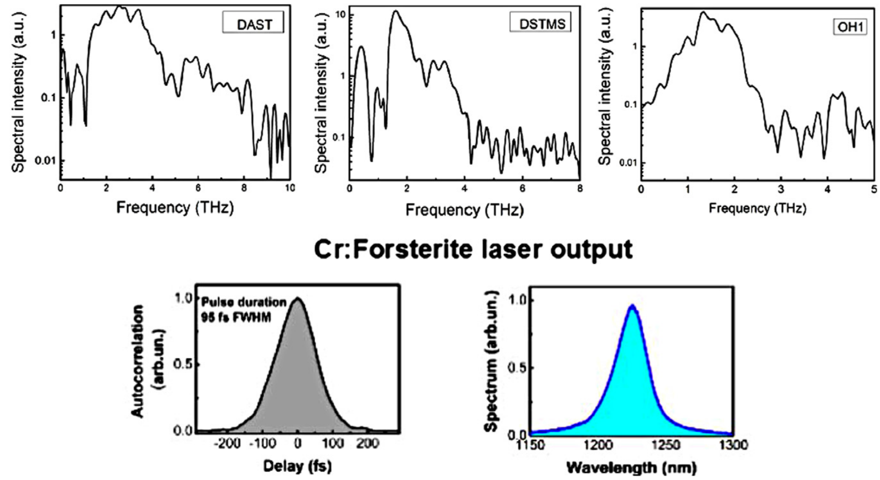

Fig. 2. Top: Terahertz spectrum emitted by DAST, DSTMS and OH1 retrieved by first order autocorrelation in a THz Michelson interferometer taken from Ref. [16]. Bottom: the Cr: forsterite laser output taken from Ref. [17].

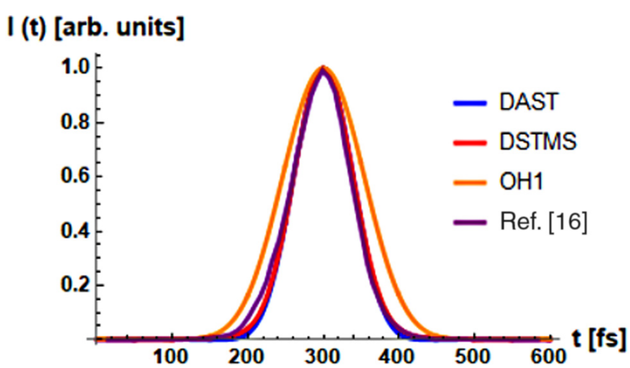

Fig. 3. The comparison between the pump envelope deduced from the autocorrelation measurement of Ref. [17] (purple curve, FWHM 95 fs) and the temporal envelopes of the pump for the different crystals, reconstructed starting from the THz spectra reported in Fig. 2. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

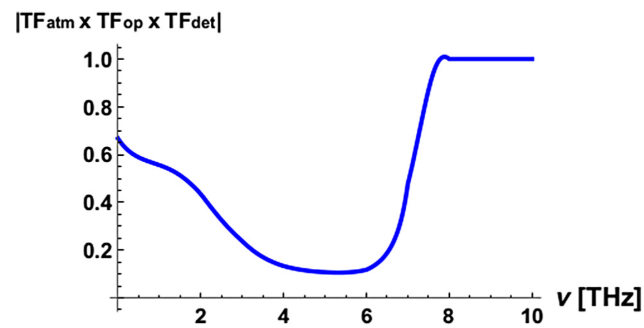

Fig. 4. Modulus of the reconstructed function $TF_{atm} \times TF_{op} \times TF_{det}$ relative to the pump retrievement of Fig. 3. This is the necessary correction to the transfer function, as expressed by Eq. (7), which allows to take into account both for the losses due to the THz propagation in air and those due to the finite acceptance and chromatic effects related to the optical components.

about the chirp and/or by higher order dispersion terms of the laser pump spectral phase. In fact we can for example consider a gaussian pump electric field:

$$E_p(\omega) = \frac{E_0 \tau}{2\sqrt{\log 2}} \cdot \exp\left[-\frac{\tau^2(\omega-\omega_0)^2}{8\log 2} + i\frac{a}{2}(\omega-\omega_0)^2 + i\frac{b}{6}(\omega-\omega_0)^3 \right] \quad (8)$$

where τ is the transform-limit FWHM, a is the chirp coefficient, b the third order dispersion coefficient, and E_0 the electric amplitude. It can be shown that the convolution in Eq. (6) for this case gives:

$$\left(E_p \star E_p^*\right)(\omega) = \frac{-2\pi i}{(3c_3)^1/3} exp\left[c_3 q + c_0\right] Ai\left(\frac{-ig}{3^1/3}\right) \quad (9)$$

where $c_3 = -(1/3)ib, c_2 = ib\omega/2 - \tau^2, c_1 = 2\tau^2\omega - ia\omega - ib\omega^2/2, q = -c_1 c_2/3c_3^2 + (2/27)c_2^3/c_3^3, c_0 = -\tau^2\omega^2 + ia\omega^2/2 + ib\omega^3/6, p = c_1/c_3 - c_2^2/3c_3^2$ and $g = c_3^{2/3} p$. The notation for the Airy function of first kind $Ai(x)$ has been adopted. The expressions given in Eqs. (6) and (8) hold exactly in the gaussian case. Therefore from Eq. (9) it is possible to see that the spectral phase of the pump pulse is transferred to the THz pulse.

This characteristic is important because it allows to retrieve, with a procedure similar to the one discussed above, not only the pump temporal intensity but also the pump spectral phase [20]. This is done by a coherent measurement of the THz electric field (for example via the standard Electro-Optical-Sampling technique). It is important to underline that the knowledge of the bandwidth of the pump spectral intensity is an important parameter required to remove the uncertainty related to the fact that a transform limit pump produces the same THz field that a chirped pulse with the same temporal length as the transform limited one would produce [20].

4. THz shaping by non linear phase modification

The properties shown in Eq. (9) express the fact that the pump spectral phase is transferred into the THz phase. This effect, can be used to perform the shaping of the THz waveform as shown by numerical examples in Figs. 5 and 6. In particular in Fig. 5 we plotted the THz field generated with different value of the chirp coefficient (*"a"*) while in Fig. 6 we show the THz generation for different values of the third order phase term (*"b"*). This effects could be used to properly adapt the THz waveform according to the experimental goal and can be extended to the case when both of the coefficients are changed. The modulation of the waveform result also in a decrease of the peak of the THz electric field.

5. Conclusion

In conclusion we have introduced a non-intercepting, single-shot technique to retrieve the temporal intensity profile and spectral phase of ultra-short pump lasers (100 fs and below) used to produce THz pulses

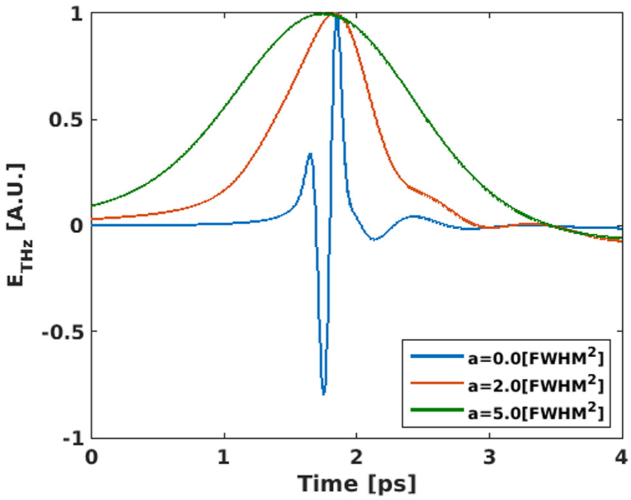

Fig. 5. THz field produced by OR in DSTMS crystal $L = 0.5$ mm with a pump of $\lambda = 1250$ nm and FWHM = 100 fs for different value of the chirp coefficient a. The amplitudes have been normalized to obtain the same peak value for easier reading.

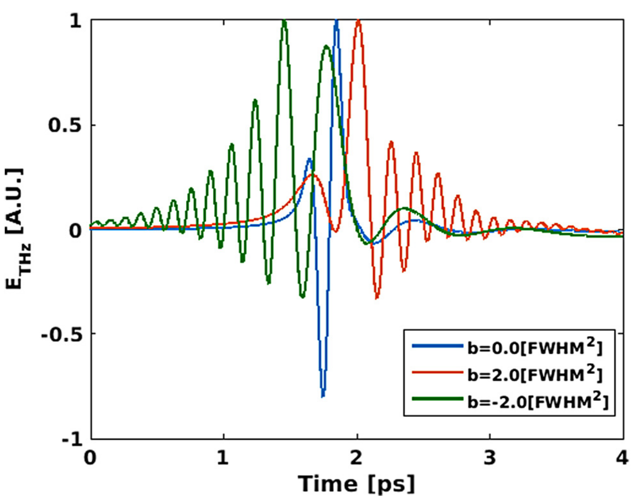

Fig. 6. THz field produced by OR in DSTMS crystal $L = 0.5$ mm with a pump of $\lambda = 1250$ nm and FWHM = 100 fs for different value of the third order dispersion coefficient b. The amplitudes have been normalized to obtain the same peak value for easier reading.

via OR. We have shown that the pump spectral phase is transferred to the THz domain and that this property can be used for the shaping of THz waveform. This is an important aspect of great interest for many applications, including the most recent ones related to electron acceleration experiments [12]. Therefore this work paves the way to the possibility to coherently and dynamically control the THz field shape.

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