Destructive interference of freely propagating terahertz pulses and its potential for high-resolution spectroscopy and optical computing

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Freely propagating terahertz pulses have been superimposed on a detection crystal leading to their mutual annihilation. An extinction ratio as good as 10:1 is found. The interference pattern represents the sum of the pulses, as measured individually, with high fidelity. Its application for high-resolution spectroscopy and optical computing are discussed. © 2001 American Institute of Physics. [DOI: 10.1063/1.1381419]

Far-infrared spectroscopy experienced increased activity lately due to the development of techniques for the generation and detection of high-intensity, freely propagating electromagnetic pulses generally known as terahertz (THz) pulses. These have been applied to study a wide variety of properties of gases, liquids, solids, and were used, e.g., for imaging and ranging applications. In this letter we present a THz-spectroscopy technique based on the coherent superposition of two similarly generated THz pulses caused—dependence of their relative phase and polarity—mutual annihilation or amplification. This constitutes an educative example of the electromagnetic properties of these pulses and has potentially important applications for all-optical signal processing and high-resolution spectroscopy.

For our experiments we used a home-built femtosecond oscillator providing pulses of approximately 5 nJ with a pulse duration (after compression) of 22 fs at 790 nm center frequency and 80 MHz repetition rate. These pulses were split into four beams of different relative intensity as indicated in Fig. 1: approximately 2% of the pulse energy was used to monitor the spectrum and stability of the oscillator during the measurement (not shown); 10% of the original beam (dotted line) were used for detection of the THz wave form; the remainder was split into two even parts (solid and dashed lines) for the generation of two independent, counter-propagating THz pulses.

As indicated in Fig. 1, one of these pulses (solid line) was brought onto the delay line D2 and subsequently was guided through a small central hole in a parabolic mirror onto a ZnTe crystal [0.5×5×5 mm, (110) oriented, supplied by Superconix]. This leads to the generation of a THz pulse which is subsequently chopped at approximately 900 Hz. Then the pulse is collimated by a parabolic mirror. After collimation it can be passed through a sample cell (not done in the shown experiments). Finally it is deflected by two plain, aluminum-covered mirrors and focused onto a ZnTe crystal identical to the one used for its generation. By electro-optic sampling its electric field is converted to a intensity signal by use of two crossed polarizers. This signal is acquired by a photodiode and lock-in technique is applied. A thin, black membrane is placed before the final focusing mirror in order to block the 790 nm light of the generation pulse from passing along with the THz pulse to the detection crystal where it would cause a background signal detrimental to the experimental resolution. Scanning the delay line D1 the temporal wave form of the THz pulse can be acquired. A typical wave form is shown as each of the thin lines of Fig. 2. This letter describes the phenomena which can be observed, if a second THz pulse is brought simultaneously onto the same detection crystal.

The dashed lines in Fig. 1 indicate the path taken by the second main pulse. It is initially delayed (D2) and then

FIG. 1. Experimental setup: initially the probe pulse (dotted line) is split off from the incoming pulse (bottom right). The remainder is split in two even parts (solid and dashed line). Both pulses are guided antiparallelly onto a ZnTe crystal for THz generation. The resultant THz beams are collimated by parabolic mirrors, passed through an area where a sample/reference cell can be placed (no sample cells were used in the measurements presented here) and are subsequently focused on a detection crystal. Here their electric field is converted to an amplitude modulation of the probe pulse using a combination of crossed polarizers and a ZnTe crystal.
passed through a small hole in the parabolic mirror onto the same spot of the generation crystal. The generated THz pulse is also collimated by a parabolic mirror, and subsequently guided by two flat mirrors before it is focused onto the same spot of the detection crystal as the first one. The two flat mirrors in the center of the setup are stacked vertically. The THz pulses of the two arms propagate above and parallel to each other on the center stretch before being focused by the parabolic mirror on one spot. For the measurements shown, the whole setup of Fig. 1 is purged with dry nitrogen.

The THz pulses are generated at a single spot of the generation crystal using beams propagating antiparallelly. When the generation crystal is rotated correctly, this produces two pulses with identical wave form and inverted sign\(^{10}\) (see thin lines in Fig. 2). In the present setup the generation of the two THz pulses occurs slightly time delayed, as the arms in which the THz pulses propagate are 2 cm different in length. However, no effects of the interference of pump pulses were observed using arms of identical length. This can be understood realizing that interference components cannot be phase matched during the propagation of the pump pulses through the generation crystal. In the detection crystal both THz pulses arrive with a delay set by D2. If it is adjusted to no phase difference between them, then they interfere destructively, thus causing the annihilation of the measured signal in the detection crystal (thick line in Fig. 2).

The achievable reduction of the THz signal is solely limited by the degree of identity of the pulses and the timing precision; the interference wave form is within experimental error identical to the mathematical difference of the wave forms of the two individual pulses. The spectrum of the mathematically created and the measured interference pulses are virtually identical on the scale of the plot and both differ significantly from the spectrum of each of the individual pulses (top inset in Fig. 2). The data shown in Fig. 2 corresponds to an extinction ratio of approximately 10:1 between the original pulse and the destructive interference pattern. This corresponds to a base line reduction by 90% in a single measurement without requiring individual runs for sample and reference each. Note that the presented wave forms are single scans; no averaging was applied. We attribute the remaining signal to (a) imperfections in the imaging system, (b) to the length difference of approximately 2 cm between both arms causing slightly different absorption along the beam path albeit extensive purging with N\(_2\), (c) to the limited ability to split the incoming beam into two exactly identical parts, and (d) to the limited resolution of the delay stage D2 allowing to match the offset of the pulses no better than by 1 \(\mu m\). The latter is believed to be the major source of the small ripples found in the temporal trace. In principle, it could be farther reduced using piezoactuated delay stages. The magnitude of the THz pulses is fine tuned by the precise orientation of the generation crystal, which allows for very good matching of the pulse amplitudes. The difference in length between the arms (b) is necessary in order to prevent effects from backreflection of the pump beam in the generation crystal: about 30% of the light transmitted through the ZnTe crystal is actually reflected on its backsurface causing THz generation on its return through the crystal. The thus created pulse would be superimposed on the observed wave forms if no length difference between the two arms were chosen. This effect is, however, only due to the use of a single generation crystal in our setup and is no fundamental limitation of the method. Similar to destructive interference, also constructive interference can be induced by setting a delay of \(\frac{1}{2}\lambda\) using D2.

Two application areas of the method presented are evident: (a) signal processing and (b) high-resolution spectroscopy. Concerning (a) it has to be pointed out that the entire setup constitutes a purely optical XOR gate for near-infrared pulses; while the logic operation of signal extinction (if inputs arrive at both arms simultaneously) or transmission (if either of them arrives individually) is performed with THz pulses, their generation is performed using near-IR pulses and they are transcribed onto a near-IR pulse at the end of the experiment. Thus, as a whole the setup operates as an XOR gate for near-IR pulses. While in our study 790 nm pulses were employed, similar results are to be expected using the telecommunication-relevant wavelengths in the near IR. The signal-to-background contrast achievable by electro-optic sampling has been proven to be sufficient for the acquisition of a whole THz wave form using a single laser shot.\(^{11}\) For the detection of the logic information here only one point of the wave form is sufficient. This is significantly easier than what has been achieved previously. Indeed, we could lately realize THz wave form acquisition similar to Ref. 11 using our nonamplified system.\(^{12}\) Given the duration of the pulse of significantly less than 10 ps, logic operations on the 100 GHz scale are possible in this setup, sufficient for all presently relevant technologies.

THz gas-phase spectroscopy suffers greatly in its applicability as high-resolution spectroscopic technique from the small absorption strength of thin sample volumes. Here our method of inherent background subtraction can provide valuable help. Small variations between two pulses can be detected very sensitively by the method presented here. It is

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**FIG. 2.** The main plot shows two antisymmetric THz wave forms (thick lines) and their superposition (thick line) as acquired by electro-optic sampling in a ZnTe detection crystal. The bottom insert is a magnification of the area of maximum electric field. The top insert shows the frequency spectrum of the measured interference pattern in comparison to the spectrum of a single pulse. All traces shown are single scans acquired at a scan speed of approximately 1 ps of delay per minute; no averaging or smoothing was applied.

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Notes:

1.~Ref. 11 using our nonamplified system.\(^{12}\) Given the duration of the pulse of significantly less than 10 ps, logic operations on the 100 GHz scale are possible in this setup, sufficient for all presently relevant technologies.
especially suitable to monitor small absorption differences between a sample volume and a reference cell; large differences are not so easy to measure as they generally go along with significant phase shifts of frequency components close to the investigated absorption line. This prevents complete destructive interference. An additional advantage of this method over the conventional subtraction of subsequently acquired sample and reference spectra is the automatic compensation for laser power fluctuation, as these effect both arms similarly.

In summary, we present evidence of mutual annihilation of freely propagating THz pulses. Constructive interference of superimposed pulses can similarly be achieved. Applications for signal processing and spectroscopic purposes are discussed.

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