Direct field-resolved detection of terahertz transients with amplitudes of megavolts per centimeter

K. Reimann, R. P. Smith,* A. M. Weiner,† T. Elsaesser, and M. Woerner

Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Strasse 2A, 12489 Berlin, Germany

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Phase-matched difference-frequency mixing in a thin GaSe crystal within the broad spectrum of 25-fs pulses from a Ti:sapphire oscillator multipass amplifier system permits the generation of few-cycle electric field transients, frequencies up to 30 THz, and amplitudes of more than 1 MV/cm. The field transients generated at a 1-kHz repetition rate are directly measured by electro-optic sampling by 12-fs probe pulses from the 75-MHz repetition-rate Ti:sapphire oscillator in combination with a novel electronic gating technique. © 2003 Optical Society of America

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Terahertz (THz) generation and spectroscopy have developed into an important field of ultrafast science.[1,2] Early research concentrated on the generation of picosecond pulses in a frequency range up to approximately 3 THz. These pulses have been used mainly for measuring steady-state THz spectra, imaging, and microscopy and—to a lesser extent—as probe pulses in ultrafast time-resolved experiments. More recently, optical rectification and phase-matched difference-frequency mixing of sub-15-fs near-infrared pulses at megahertz repetition rates have allowed for the generation of electric field transients of sub-200-fs duration with frequencies up to ~50 THz.[3–5] Such transients have been fully characterized in amplitude and phase by electro-optic sampling.[6–8] Although these THz transients have been applied for probing transmission changes in ultrafast experiments and have even been used to induce nonlinear excitation,[9] their field amplitude of ≤10 kV/cm is comparatively small. Much higher field amplitudes, in the megavolt-per-centimeter range, have been achieved by difference-frequency mixing of pulses derived from amplified Ti:sapphire lasers working at kilohertz repetition rates.[10,11] Here, however, the THz pulse duration is typically several hundreds of femtoseconds, i.e., many optical cycles, and, even more important, full characterization with respect to amplitude and phase is still lacking.

In this Letter we report a new scheme for the full characterization of THz transients with megavolt-per-centimeter amplitude and a 1-kHz repetition rate. Pulses of 25-fs duration from an amplified Ti:sapphire laser system serve for phase-matched difference-frequency mixing in thin GaSe crystals. The generated field transients are directly measured by ultrafast electro-optic sampling in a 10-μm-thick (110)-oriented ZnTe crystal by use of 12-fs probe pulses from the Ti:sapphire oscillator working at a repetition rate of 75 MHz in combination with a novel electronic gating technique. A 30-μm-thick GaSe crystal permits the generation of an electric field transient with only 1.3 cycles (number of cycles within the FWHM of the intensity transient) that covers a broad band of frequencies up to 35 THz and has an electric field strength of 1 MV/cm. For thicker GaSe crystals one observes more optical cycles of similar amplitude (longer pulses with more energy) and some tunability by changing the phase-matching angle.

The experimental setup is shown in Fig. 1(a). A Ti:sapphire oscillator multipass amplifier system (Femtolasers Femtopower Compact Pro) working at a 1-kHz repetition rate generates nearly Fourier-limited pulses centered at a wavelength of 793 nm with a bandwidth of 43 nm (FWHM) and a duration of 25 fs. Pulses of typically 600-μJ energy propagate unfocused (beam diameter, 7 mm) through GaSe crystals of various thicknesses, in which ultrafast electric field transients with frequencies up to 35 THz are generated by phase-matched type I difference-frequency mixing.[4]

For a complete characterization of the THz transients, we perform a direct time-resolved measurement of the electric field by electro-optic sampling.[6] After passing through a thin germanium beam splitter (high transparency and low dispersion) the ultrafast electric-field transient is focused by a 90° off-axis parabolic mirror with 12.7-mm focal length onto a 10-μm-thick (110)-oriented ZnTe crystal. At the germanium beam splitter the near-infrared probe beam is combined with the THz beam and focused onto the same position of the ZnTe crystal. The electro-optic modulation of the 12-fs-long probe pulse induced by the ultrafast Pockels effect is detected with a pair of fast balanced photodiodes (Fig. 1, PD1 and PD2; Centronic AEPX65). By varying the time delay between the THz and the near-infrared pulses we directly sample the temporal waveform of the ultrafast electric field transient. For a typical transient the signal at every time delay is averaged over 20 shots and the time delay is scanned with a step size of 0.4 fs.

Because the probe beam is derived (beam splitter BS; reflection, 20%) from the Ti:sapphire oscillator, which has a repetition rate of 75 MHz, only one pulse in 75,000 coincides with an amplifier pulse. To extract the pure electro-optic signal induced by the electric field of the THz pulse we use the novel electronic gating technique illustrated in Fig. 1(b). The difference signal PD1 − PD2 passes through two electronic gates (Stanford Research SR250). Gate 1 extracts the

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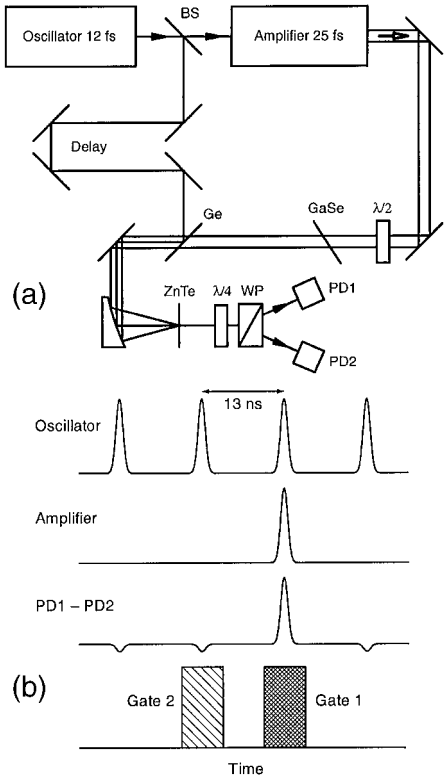

Fig. 1. (a) Setup for the generation and direct field-resolved detection of high-field THz transients with frequencies up to 35 THz: WP, Wollaston polarizer. The transients are generated by phase-matched difference-frequency mixing in GaSe of intense 25-fs pulses from an amplified Ti:sapphire laser. The electric field is directly measured by electro-optic sampling in ZnTe with 20% of the oscillator beam used as 12-fs probe pulses. (b) Schematic showing the timing of various electric signals that occur in the novel gating technique: 75-MHz train of oscillator pulses (upper trace), amplified pulses at a 1-kHz repetition rate (trace below the upper trace), electronic difference signal from the balanced photodiodes (next lowest trace), and electronic gates for signal (Gate 1) and reference (Gate 2) (bottom trace).

signal from the oscillator pulse in coincidence with the THz transient and Gate 2 measures a reference signal by using the previous oscillator pulse. For each single shot this reference is subtracted from the actual signal before averaging. In this way we eliminate deviations from a perfect intensity balance between PD1 and PD2, which typically occur as small drifts on a time scale of minutes.

In Fig. 2(a) we show an electric field transient (solid curve) generated in a 200-μm-thick GaSe crystal for an external phase-matching angle of 44°. The difference signal from the two balanced photodiodes (left axis) is plotted as a function of the time delay between the 12-fs oscillator pulses and the THz transient. We define the maximum of the transient as delay zero. The time evolution of the electric field [Fig. 2(a)] with a period of 60 fs is in excellent agreement with the prediction of the theoretical model for phase-matched difference-frequency mixing.[5] Transients with a similar time evolution were generated with pulses from high-repetition-rate oscillators.[6,7] The electric field strength of the present pulses, however, is orders of magnitude higher than in any transient generated with oscillator pulses. There are two ways to determine experimentally the absolute value of the electric field: On the one hand, the electric field can be obtained from the pulse energy of 12 nJ measured with a bolometer, from the size of the beam waist of 50 μm (FWHM), and from the pulse duration as measured by the electro-optic signal. On the other hand, one can use the absolute value of the electro-optic signal to calculate the electric field, using the electro-optic coefficient of ZnTe (3.9×10^{-10} cm/V),[12] the transmission of the electric field through the ZnTe surface, the frequency-dependent sensitivity of the electro-optic sensor that is due to the group-phase velocity mismatch, and the finite duration of the probe pulses.[13] The two methods agree within experimental error, to yield very high amplitudes of the electric field, of the order of 1 MV/cm [right-hand ordinate scales in Figs. 2(a) and 2(b)].

In Fig. 2(b) we show transients obtained with a 30-μm-thick GaSe crystal. Interestingly, the $\chi^{(2)}$ nonlinearity directly shows the $\bar{6}$ symmetry of the GaSe crystal along its c axis (in contrast, the linear optical properties for light incident along c are isotropic). As an example, after a 180° rotation of the GaSe crystal about its c axis the emitted field reverses its absolute sign [dashed curve in Fig. 2(b)].

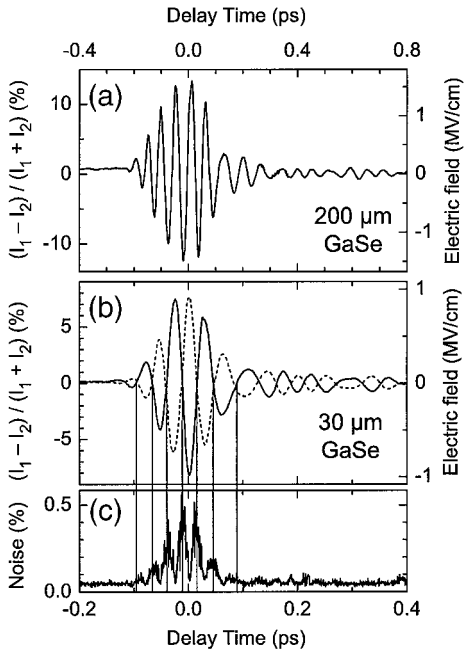

Fig. 2. (a) Electric field transient (solid curve) generated in 200-μm GaSe for an external phase-matching angle of 44°. The normalized difference signal from the two balanced photodiodes (left axis) is plotted versus the time delay between the 12-fs-long oscillator pulses and the THz transient (right axis, corresponding electric field strength). (b) Solid curve, transient obtained for 30-μm-thick GaSe. Dashed curve, the field changes its absolute sign after a 180° rotation of the GaSe crystal about its c axis. (c) The noise (rms of single-shot fluctuations) shows delay fluctuations between oscillator and amplifier pulses of less than 1 fs rms.

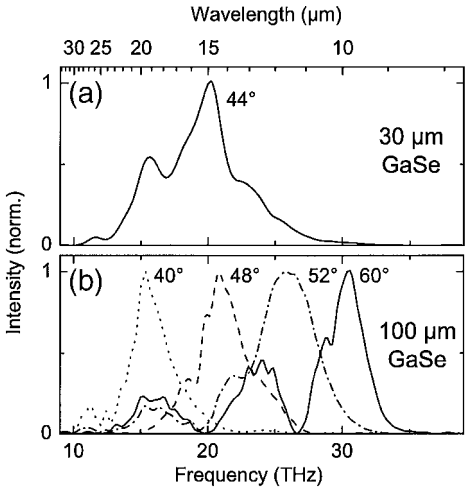

Fig. 3. Spectra (obtained by Fourier transform of the measured transients) (a) for a 30-μm-thick and (b) for a 100-μm-thick GaSe crystal for several external phase-matching angles as indicated.

In our detection scheme the accuracy of the time-resolved measurement of the THz field is limited by a possible timing jitter between the oscillator pulses and the amplifier pulses. A detailed analysis of the noise of the electro-optic signal allows us to determine such timing fluctuations. In Fig. 2(c) the noise (rms of single-shot fluctuations) of the electro-optic signal is plotted as a function of time delay. The noise consists of a background level that is caused mainly by shot noise as a result of the limited number of photons in the probe pulses and by additional noise during the THz transient. The noise shows maxima at the nodes of the electric field transient and minima at the antinodes, as expected for averaging of the momentary THz field over a narrow time interval defined by the jitter. From the ratio of noise amplitude to slope at the nodes one can estimate the rms jitter to be less than 1 fs, which is much smaller than the pulse duration of the Ti:sapphire oscillator. As the noise at the antinodes of the THz transients is not significantly above the background level, we can conclude that the shot-to-shot amplitude fluctuations of the electric field are quite small ($\approx 10^{-3}$). Interestingly, they are considerably smaller than the fluctuations of the amplified pulses ($\approx 10^{-2}$). This effect may be caused by the onset of two-photon absorption in the GaSe crystal, which decreases the efficiency of difference-frequency generation for high intensities. On a time scale of minutes and longer we observe additional weak timing drifts ($\approx$10 fs), which are probably due to thermal fluctuations of the whole experimental setup.

It is worth comparing the present results with data obtained by use of the amplified pulses as probes. First, the amplified pulses are longer than the oscillator pulses (25 compared with 12 fs), which reduces the signal (at 10 μm) by a factor of 5.[13] Second, the amplified pulses have much higher fluctuations, resulting in noise that is more than a factor of 40 higher. The two effects together yield a signal-to-noise ratio at least 2 orders of magnitude lower than that with the oscillator.

A final comment should be made on the spectra of the THz transients in Fig. 3. For thick GaSe crystals ($\geq$100 μm) the phase-matching bandwidth is smaller than the total bandwidth of the amplified near-infrared pulses, and thus one can tune the frequency position of the THz output by tilting the nonlinear crystal. For thin GaSe crystals, phase matching occurs over the full bandwidth of the pump pulses, resulting in the broad spectrum of Fig. 3(a). The spectra are in excellent agreement with calculations[5,13] that take into account phase matching (this causes the minima in the spectra for 52° and 60°), propagation of input and output pulses, and the bandwidth of the pump pulse (this limits the maximum frequency).

In conclusion, we have reported what is to our knowledge the first direct field-resolved detection of THz field transients with megavolt-per-centimeter amplitudes. Our highly sensitive method will pave the way for amplitude- and phase-resolved experiments in which strong THz fields induce a nonlinear optical response beyond the perturbative regime.

K. Reimann's e-mail address is reimann@mbi-berlin.de.

*Permanent address, Georgia Institute of Technology, Atlanta, Georgia 30332.

†Permanent address, School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907.

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