Controlled shaping of ultrafast electric field transients in the mid-infrared spectral range

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We experimentally demonstrate amplitude and phase shaping of femtosecond mid-infrared pulses in a range centered about 14 μm. Single pulses with a tailored optical phase and phase-locked double pulses are generated by phase-matched difference-frequency mixing in a GaSe crystal of near-infrared pulses shaped with a liquid-crystal modulator. The electric field transients are directly measured by free-space electro-optic sampling, yielding pulse durations of 200–300 fs. Our data are in good agreement with a model that describes phase-matched optical rectification. © 2000 Optical Society of America

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Ultrashort electric field transients with frequencies up to the mid-infrared (MIR) spectral range provide a broad potential for the study of the ultrafast dynamics of elementary excitations in condensed matter.[1] The ultimate goal of such investigations is not only to initiate and probe ultrafast dynamics, e.g., of vibrational transitions in molecules or intersubband transitions in semiconductor nanostructures, but to control coherently the dynamics of such elementary excitations in a defined manner.[2] Achieving these ends requires amplitude and phase control of the excitation pulses in their respective spectral ranges.

In recent years, experiments in coherent control have found been of increasing interest, in particular for improving our understanding of chemical reactions[3] and dynamic processes in semiconductor quantum structures.[4] Many fundamental processes in condensed matter involve excitations at energies that correspond to the MIR spectral range ($\lambda = 3–20$ μm). So far, however, it has not been possible to carry out coherent control experiments in this range because a suitable light source for shaped MIR pulses is still lacking. In this Letter we experimentally demonstrate what is to our knowledge the first controlled shaping of high-repetition-rate ultrafast electric field transients in the MIR.

The experimental setup is shown schematically in Fig. 1(a). Near-infrared (NIR) pulses centered at a wavelength of 780 nm with a large bandwidth of 70 nm are generated in a cavity-dumped, mode-locked Ti:sapphire oscillator.[5] In our setup, shaped MIR pulses are obtained in a two-stage scheme: First the beam is passed through a programmable pulse shaper, which consists of a liquid-crystal modulator array placed in a zero-dispersion stretcher unit.[6,7] In this way, amplitude- and phase-shaped (NIR) pulses are generated. Although here we have used the pulse shaper to produce phase-locked pulse pairs, it is clear that much more complex NIR pulses can be generated in exactly the same setup. In the second step, phase-matched difference-frequency mixing of the various spectral components of these pulses in a 0.5-mm-thick GaSe crystal provides femtosecond MIR electric field transients. The details of crystal orientation, polarization of the input pulses for type I phase matching, and theoretical calculations that demonstrate femtosecond MIR generation in the 7–20-μm range are described in Ref. 8.

In general complete characterization of MIR pulses is a difficult task. Standard techniques such as

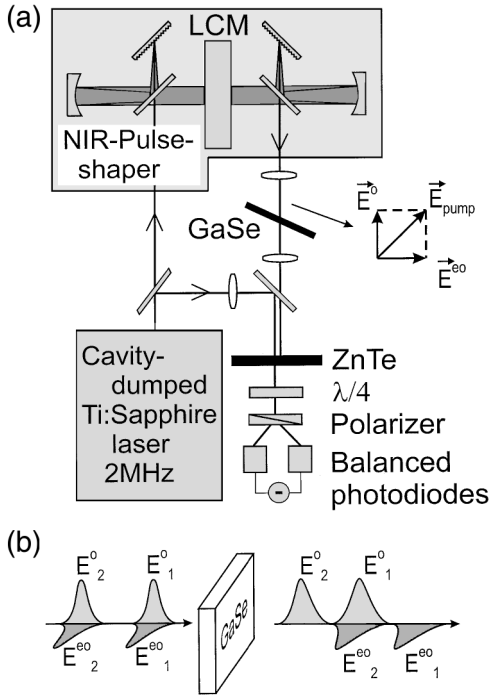

Fig. 1. (a) Experimental setup for generation and characterization of amplitude- and phase-shaped femtosecond pulses in a frequency range up to 40 THz. The ultrafast electric field transients are generated by phase-matched difference-frequency mixing in a GaSe crystal of shaped NIR pulses by use of a liquid-crystal modulator (LCM). The electric field transients generated are directly measured by ultrafast free-space electro-optic sampling. (b) Schematic showing the propagation of the ordinary (*o*) and the extraordinary (*eo*) components of two phase-locked NIR pump pulses. Group-velocity mismatch between the ordinary and the extraordinary waves leads to coincidence of the extraordinary component of the second pulse with the ordinary component of the first.

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upconversion and interferometric electric field correlation measure either the intensity envelope or the power spectrum.[8] Those techniques, however, do not provide a full characterization of the pulse; its chirp and the absolute phase remain undetermined. Complete characterization is possible only by direct time-resolved measurement of the electric field. Here we apply free-space electro-optic sampling[9] to measure the shaped MIR electric field transients.[10] For this, the shaped MIR pulse is focused onto a 10-μm-thick ZnTe crystal. An approximately 17-fs NIR reference pulse from the Ti:sapphire oscillator is focused onto the same position. The electro-optic modulation of the reference pulse induced by the ultrafast Pockels effect is detected with a pair of balanced photodiodes. By varying the time delay between the MIR and the NIR pulses, we directly sampled the temporal waveform of the MIR electric-field transient.

In Fig. 2(a) a typical MIR waveform is shown that was generated by a single (i.e., unshaped) NIR pump pulse. The corresponding spectrum as obtained by Fourier transformation of the waveform is centered about 22 THz (i.e., $\lambda = 13.7$ μm) with a FWHM spectral width of approximately 2 THz, as shown in the inset. We then imposed a linear spectral phase $\Phi(\omega)$ on the NIR pulse, which delayed the NIR and consequently the MIR field transients by 60 fs, as shown in Fig. 2(b). Next we programmed the pulse shaper to produce a pair of in-phase NIR pump pulses with 60-fs separation. The resulting MIR transient is shown in Fig. 2(c). In addition to the transients generated by the two pulses individually, i.e., the expected superposition of the transients in Figs. 2(a) and 2(b), we find a second pulse, centered about 400 fs, well after the maxima of the two phase-locked pulses. Similar results were obtained when the phase-matching angle was changed to yield a MIR centered at 12.5 μm. To explore this phenomenon further, we used the pulse shaper to create NIR pulse sequences with pulse separations of exactly one half or one oscillation period at the 22-THz carrier frequency of the generated MIR light. In Fig. 3 we plot the electric field transients generated by a pair of phase-locked pump pulses separated by $\Delta t = 23$ fs [Fig. 3(a); equal to one half of the MIR period] and $\Delta t = 46$ fs [Fig. 3(b); equal to a full MIR period]. Solid and dashed curves correspond to a relative phase $\Delta\Phi$ between phase-locked pump pulses of $\Delta\Phi = 0$ and $\Delta\Phi = \pi$, respectively. The inset shows an enlarged time window near 400 fs in which the wave form generated shows the following interesting behavior: The amplitude depends only weakly on the coarse separation between the phase-locked pump pulses, whereas the phase changes by π as we vary the relative phase between the pump pulses by the same amount. In contrast, in the time range near delay zero we obtain the expected behavior: (i) we observe either destructive [Fig. 3(a)] or constructive [Fig. 3(b)] interference between the two generated MIR fields and (ii) the phase of the electric field transients is independent of the relative phase between the pump pulses as expected for optical rectification. We explain the peculiar behavior observed in our experiments as follows: At the entrance of the GaSe crystal we have two pump pulses, $E_1(t)$ and $E_2(t)$ [$=E_1(t + \Delta t)$], each consisting of an ordinary $\mathbf{e}_o$ and an extraordinary $\mathbf{e}_{eo}$ component, i.e., $\mathbf{E}_1(t) = E_1(t)[\mathbf{e}_o + \mathbf{e}_{eo}]/\sqrt{2}$ and $\mathbf{E}_2(t) = E_2(t)[\mathbf{e}_o + \mathbf{e}_{eo}]/\sqrt{2}$ [see Fig. 1(a)], separated by time Δt (details of the polarization geometry are explained in Ref. 8)]. The two pump pulses generate the nonlinear polarizations: $P_1^{\mathrm{NL}}(t)\mathbf{e}_o = \mathrm{Re}\{\chi^{(2)}{:}E_1(t)\mathbf{e}_o{:}[E_1(t)\mathbf{e}_{eo}]^*\}$, $P_2^{\mathrm{NL}}(t)\mathbf{e}_o = \mathrm{Re}\{\chi^{(2)}{:}E_2(t)\mathbf{e}_o{:}[E_2(t)\mathbf{e}_{eo}]^*\}$ via the optical rectification part of the $\chi^{(2)}$ tensor. Applying these pump pulses

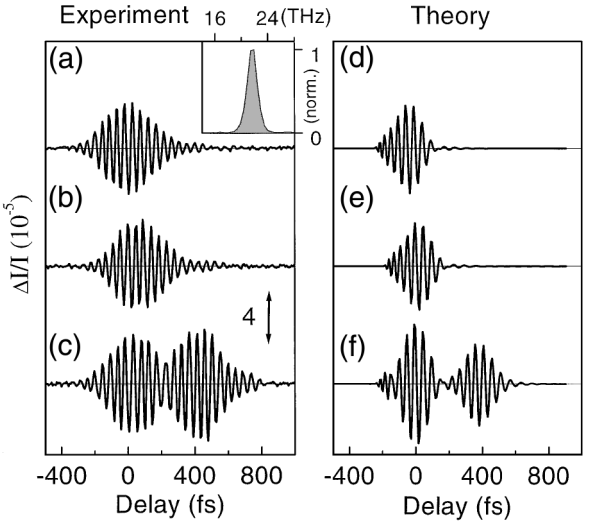

Fig. 2. Electric field transients of MIR pulses measured with free-space electro-optic sampling. The pulses centered about $\lambda = 13.7$ μm are generated by a pair of phase-locked pump pulses with 60-fs separation. (a), (b) Transients generated when each pump pulse is applied separately. (c) Applying two pump pulses. Inset, Fourier transform of the waveform of a single pulse, showing the pulse spectrum. (d)–(f) Corresponding theoretical curves from the model discussed in Ref. 8.

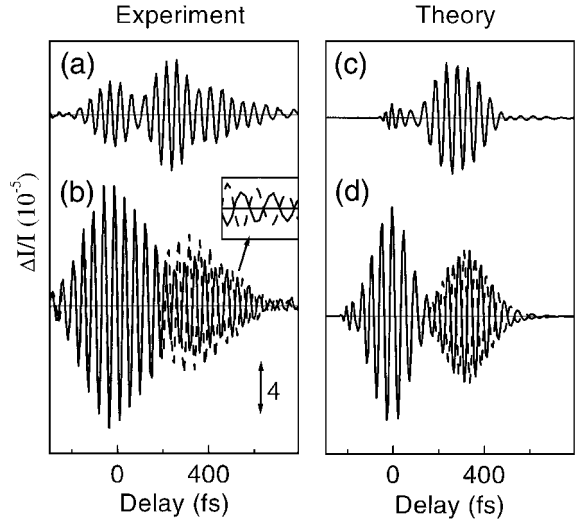

Fig. 3. MIR waveforms generated by two NIR pulses with distances of (a) $\Delta t = 23$ fs (corresponding to a $\lambda/2$ retardation of the MIR phase) and (b) $\Delta t = 46$ fs. The relative phase between phase-locked NIR pump pulses, $\Delta\Phi$, is 0 (solid curves) and π (dashed curves). Inset, enlarged time window near 300 fs. Corresponding model calculations are shown in (c) and (d).

separately, we obtain the corresponding MIR field transients [Figs. 2(a) and 2(b)]. Now, as we apply the double pulse $E_1(t) + E_2(t)$, the nonlinear polarization $P_1^{\mathrm{NL}}(t) + P_2^{\mathrm{NL}}(t) + P_X^{\mathrm{NL}}(t)$ contains in addition to the self-terms a cross term $P_X^{\mathrm{NL}}(t)\mathbf{e}_o = \mathrm{Re}\{\chi^{(2)}{:}E_1(t)\mathbf{e}_o{:}[E_2(t)\mathbf{e}_{eo}]^* + \chi^{(2)}{:}E_2(t)\mathbf{e}_o{:}[E_1(t)\mathbf{e}_{eo}]^*\}$, depending on the electric fields of both pulses.

Close to the entrance of the GaSe crystal the self-terms dominate and thus generate two MIR pulses separated by the same delay as the corresponding pump pulses [Figs. 1(b)]. In this early stage of the generation process, the cross term vanishes owing to the poor temporal overlap between $E_1(t)$ and $E_2(t)$. For situations in which Δt is below the MIR pulse duration, the two self-terms merge into one pulse, and constructive or destructive interference of the MIR fields takes place according to the specific Δt. This effect explains the electric field transients near delay zero in Figs. 2(c), 3(a), and 3(b).

The wave forms [Fig. 3(b)] observed near 400 fs, however, are caused by the cross terms. As the group velocities of the extraordinary and the ordinary pump pulses are different in GaSe, the extraordinary part of the second pump pulse meets the ordinary part of the first pump pulse at a later position in the crystal, depending on the distance between the pump pulses. Here, the contribution that is due to the respective self-term vanishes and the cross term generates the second MIR pulse near 400 fs. The large delay between the MIR pulse pair relative to that of the pump pulses occurs because the MIR/NIR group-velocity mismatch is much larger than that between ordinary and extraordinary pump waves. In contrast to the field transients generated by the self-terms, the wave form caused by the cross term is quite sensitive to the relative phase between the two pump pulses. It is obvious from the equation for P_X^{NL} that the phase of the MIR field is directly connected to the relative phase between the two pump pulses [see the solid and dashed curves in Fig. 3(b)].

Our qualitative picture is fully confirmed by detailed model calculations according to the theory derived in Ref. 8, which takes into account the full frequency-dependent refractive indices of all three interacting waves. The theoretical curves shown in Figs. 2(d)–2(f) and 3(c) and 3(d) reproduce all the main features observed in the respective experiments, as predicted by our arguments above. We attribute the relatively minor quantitative differences to a small chirp on the input pulses and to dispersion in the MIR optics between the GaSe and ZnTe crystals, neither of which is included in the theory. These results for generation of MIR pulse pairs elucidate all the essential new nonlinear optics that determine the shape of the MIR field transients.

Our results suggest that—within the constraints of the NIR spectral bandwidth—one can realize an arbitrary nonlinear polarization P_X^{NL} by exploiting the cross term between independently shaped ordinary and extraordinary input pulses. To do so would require suppression of the self-terms, which one could accomplish, e.g., by separating the two components from each other in time by passing them through a birefringent crystal before they enter the GaSe. Such a technique paves the way for generating arbitrarily shaped infrared electric field transients.

In conclusion, we have experimentally demonstrated controlled shaping of high-repetition-rate femtosecond MIR pulses. The ultrafast electric field transients are generated by phase-matched difference-frequency mixing in a GaSe crystal of NIR pulses shaped by a liquid-crystal modulator. The electric field transients generated are directly measured by ultrafast free-space electro-optic sampling. As an example, we generated various MIR waveforms by applying a pair of phase-locked pump pulses. Our experimental data are in good agreement with a model describing phase-matched optical rectification. The scheme presented here is extendable to more-complex pulse shapes and to other wavelengths and nonlinear crystals that are transparent and phase matchable within the required wavelength range. The results of this study allow us to shape ultrafast field transients in a controlled manner that can be used for coherent control experiments in the MIR wavelength range.

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