Far-infrared second-harmonic generation and pulse characterization with the organic salt DAST

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We generated the second harmonics of pulses in the far-infrared region (30–55 μm), using the organic salt dimethylamino-4-N-methylstilbazolium tosylate (DAST). We demonstrate that DAST can be used to characterize ultrashort pulses in a spectral region where no other materials are available. To illustrate the need for such characterizations, we show the effects of propagation through air on the shape of ultrashort far-infrared pulses. © 1998 Optical Society of America

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It is difficult to diagnose laser pulses in the far-infrared with nonlinear optical techniques such as second-harmonic autocorrelation measurements because conventional types of nonlinear materials possess strong absorption bands in this spectral region. This is not an issue for the characterization of terahertz pulses, which are produced with femtosecond near-IR pulses by optical rectification, because the electric field of these low-intensity pulses can be directly measured by use of a split-off near-IR synchronized pulse as a time gate. However, for far-IR free-electron lasers (FEL’s), which are capable of generating pulse energies more than 2 orders of magnitude higher than those generated by optical rectification sources, time-gated detection is not an option. This is so because no near-IR laser can be synchronized with sufficient time resolution to allow for time-gated detection of (sub)picosecond FEL pulses. Hence there exists a strong need for a nonlinear material that can be used to characterize the FEL pulses by second-harmonic autocorrelation techniques.

In recent years, new nonlinear materials have been developed that often have absorption properties that compare favorably with those of conventional nonlinear materials such as CdTe and AgGaSe₂. One of these materials is the organic salt dimethylamino-4-N-methylstilbazolium tosylate (DAST). DAST is known for its large second-order nonlinear optical susceptibility χ(2). It was previously used to generate second harmonics in the near IR and to generate terahertz radiation in the far-IR by use of visible pulses. Here we show how DAST can be used to characterize intense far-IR pulses in the wavelength region from 30 to 55 μm.

The FEL for infrared experiments (FELIX) at the Institute “Rijnhuizen,” FOM, Nieuwegein, The Netherlands, emits ultrashort tunable pulses at wavelengths between 5 and 100 μm and with energies of as much as 75 μJ per pulse. In our setup, FELIX pulses are first passed through a bandpass filter or a wire mesh to remove the higher harmonics generated in the FEL. Subsequently the pulses are split into two and are noncollinearly focused onto a DAST crystal. The (non-phase-matched) second-harmonic radiation generated between the two transmitted beams is detected with a HgCdTe detector as a function of time delay between the two pulses. A filter in front of the detector removes the scattered fundamental light of FELIX. The whole experimental setup is enclosed in a box that can be purged with dry-nitrogen gas.

DAST crystallizes in the acentric monoclinic Cc space group, point group m (standard orientation). From this structure it is expected that the nonlinear optical susceptibility χ(2) is largest along the a axis of the crystal. This was confirmed in second-harmonic generation (SHG) experiments in the near IR and in terahertz optical rectification experiments. To demonstrate this orientational dependence we mounted a DAST crystal upon a rotation stage. The second-harmonic intensity was measured as a function of the crystal orientation with respect to the polarization of the optical electric field. The results for a wavelength of 53 μm are shown in Fig. 1. The figure demonstrates that the efficiency of the SHG is enhanced by almost 3 orders of magnitude when the optical polarization is parallel to the crystal a axis compared with the situation when the polarization is orthogonal to the crystal a axis.

Figure 2 shows an autocorrelation trace of the FELIX pulses at 53 μm. Fitting the trace in Fig. 2 to the correlate of a Gaussian pulse, we obtained a pulse width of 1.50 ± 0.08 ps (FWHM). This pulse width corresponds to 8.5 cycles of the electric field at this wavelength, and the pulses are, therefore, comparable with the 6-cycle FELIX pulses at 10.4 and 24.5 μm reported earlier. We have measured autocorrelations at several fundamental wavelengths between 30 and 55 μm (not shown here), obtaining pulse durations between 1 and 2 ps. Our ~200-μm-thick DAST crystal has as much as 30% transmittance at the fundamental wavelength as well as at the second-harmonic wavelength in this wavelength range. The transmittance spectrum
contains a few dips where the transmittance drops to 10%. As long as the coherence length of the second-harmonic process is significantly shorter than the absorption length, the dips have little effect on the SHG efficiency. In principle, these dips could affect the shape of the autocorrelation measurement. However, we did not observe any effect of these absorption bands on the shape of the autocorrelation trace, probably because the width of the absorption bands (20–30 cm\(^{-1}\)) is comparable with, or slightly narrower, than the estimated pulse bandwidth (25–35 cm\(^{-1}\)) and because the absorption is generally weak. In addition, for the results shown in Figs. 2 and 3, the transmittance did not vary by more than a few percent over the estimated bandwidth of the pulses.

In Fig. 3(a) we show an autocorrelation measurement of FELIX pulses at 33 \(\mu\)m after they have propagated 3 m through air. Instead of a Gaussian-shaped autocorrelation we observe a strong beating pattern, which disappears when the setup is purged with dry nitrogen [Fig. 3(b)]. We were able to model the beating of the pulse by considering the strong atmospheric-water absorption lines in this spectral region. We calculated the pulse shape of a Gaussian pulse propagating through air, while considering the absorption and the refractive index (shown in Fig. 4) for the different frequency components. The absorbance is defined as \(A(\omega) = (2\omega\kappa/c)L\), where \(\omega\) is the frequency in wave numbers, \(\kappa\) is the imaginary part of the index of refraction, \(c\) is the speed of light, and \(L\) is the length of propagation (3 m). From Fig. 3 it can be seen that we obtain good agreement between our experimental data and the calculations. We obtain the best agreement with the measured curve by taking a Gaussian pulse with a FWHM of 1.4 ps and a small chirp of \(\varphi(t) = \exp[-2\ln(2(t/\tau_p))^2]\). This phase modulation broadens the energy spectrum of the pulse by only \(\sim10\%\) and considerably improves the fit. It affects the pulse shape strongly, because the beats are caused by the interference between a propagating field and the free-induction decay.
Fig. 4. Absorption and change in refractive-index spectra of water vapor in air used in modeling the propagation effects on a Gaussian pulse. (a) Seven absorption lines near 300 cm\(^{-1}\) are used in the model. These lines are (relative intensities are given in parentheses) 303.14 cm\(^{-1}\) (0.723), 302.99 cm\(^{-1}\) (1), 302.93 cm\(^{-1}\) (0.332), 301.85 cm\(^{-1}\) (0.240), 298.42 cm\(^{-1}\) (0.046), 290.75 cm\(^{-1}\) (0.04), and 289.43 cm\(^{-1}\) (0.425). (b) Because of the strong absorption, the refractive index varies wildly in this spectral region. The refractive index's deviation from 1 is plotted here.

We calculated that 74% of the light is absorbed, which is in good agreement with the experimentally observed value of 60 ± 10%. The absorption as well as the refractive-index variation had to be taken into account to model the data. We found that the beating is caused by both dispersive propagation (polariton effects) and absorption around only one water-vapor absorption line. Both effects are essential for the good agreement with the data shown in Fig. 3. We can understand the importance of dispersive propagation because near this particular waterline the refractive index varies wildly with frequency (Fig. 4). Therefore, different frequency components of the pulse travel at different speeds, giving rise to a propagation-distance-dependent interference, which modifies the pulse shape.

In conclusion, we have shown that DAST can be used to characterize far-IR ultrashort pulses between 33 and 50 μm. We observed that an initial Gaussian pulse can be dramatically distorted in this wavelength region. We explained this as being due to the presence of atmospheric-water absorption lines resonant with the laser spectrum, which gives rise to absorption and dispersive propagation effects, which are essential to explain the beats in our autocorrelation measurements.

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References

7. For the SHG of the fundamental wavelength between 30 and 36 μm we used a bandpass filter with a 23–35-μm transmittance window.
8. For the SHG of the fundamental wavelength between 47 and 55 μm we used a wire mesh filter, which is a long-wavelength pass filter with a cut-on wavelength of ~30 μm.
9. Anhydrous crystals of DAST were purchased from Molecular Opto-Electronics Corporation, Watervliet, N.Y.
10. A ZnSe filter (transparent for wavelengths below 20 μm) removed the fundamental wavelength between 30 and 36 μm, and the 23–55-μm transmittance bandpass filter removed the fundamental wavelength between 47 and 55 μm.