Generation of tunable subpicosecond light pulses in the midinfrared between 4.5 and 11.5 μm

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Stable subpicosecond infrared pulses in the spectral region of 4.5–11.5 μm are generated by difference-frequency mixing in $AgGaS_2$. The system uses femtosecond pulses from a Ti:sapphire regenerative amplifier and from a tunable traveling-wave dye laser. The infrared pulses have a duration of 400 fs, an energy of more than 10 nJ, and a repetition rate of 1 kHz.

The observation of vibrational modes with high time resolution has proved to yield important information on fast reactional and relaxational processes in solid-state physics, in photochemistry, and in biophysics. For the study of those fast reaction dynamics, infrared spectroscopic techniques are required that permit a time resolution much less than 10^{-12} s. There are various approaches for the generation of short pulses in the midinfrared region.

Optical parametric generation, optical parametric frequency mixing, and optical parametric oscillation in nonlinear crystals based on Nd:YAG or Nd:glass laser pulses were widely applied for the generation of high-power midinfrared pulses in the spectral range between 1 and 10 μm with pulse durations of several picoseconds.[1,2] Amplified dye laser pulses were used to achieve intense pulses in the subpicosecond regime.[3-6] Those systems used nonlinear crystals, such as $LiIO_3$, $LiNbO_3$, or beta-barium metaborate, which have a limited transmission range in the midinfrared ($\lambda < 5.5$ μm). With the advent of Ti:sapphire lasers[7] and amplifiers,[8] there exists a new powerful femtosecond light source with high stability and energy. The output of a two-color femtosecond Ti:sapphire laser was mixed in a $AgGaS_2$ crystal, obtaining subpicosecond pulses at 9 μm.[9] In this Letter we present a system based on a Ti:sapphire regenerative amplifier that generates high-power subpicosecond laser pulses tunable between 4.5 and 11.5 μm.

A schematic of the experimental setup is shown in Fig. 1. The source of ultrashort light pulses is a Ti:sapphire laser–amplifier system that delivers light pulses with a pulse duration of 120 fs at $\lambda = 815$ nm, a pulse energy of 700 μJ, and a repetition rate of 1 kHz.[8] The light pulses are used for (i) the generation of tunable near-infrared pulses and (ii) difference-frequency mixing. The Ti:sapphire amplifier delivers sufficient energy to pump several difference-frequency generation processes in parallel.

For the generation of stable and intense midinfrared pulses, a powerful light source is necessary that delivers tunable red-shifted laser pulses synchronized to the Ti:sapphire pump pulses. The projected tuning range of 4.5–11.5 μm in combination with the wavelength of the pump pulses (815 nm)

requires a smooth tuning of the near-infrared pulses between 870 and 1000 nm. The use of a traveling-wave dye laser[10] (TWDL) to generate intense tunable light pulses was reported previously.[5]

A single-stage TWDL emits femtosecond pulses at the long-wavelength absorption edge of the laser dye with a spectral bandwidth of approximately 30 nm. The central wavelength is fixed by the choice of the laser dye. Since there are only a few laser dyes available in the region of interest (between 870 and 1000 nm), the system should be tunable.[11,12] For this purpose we have developed a two-stage traveling-wave dye laser–amplifier configuration (see Fig. 2). Approximately 15 μJ of the amplified Ti:sapphire pulses are focused by a 6-cm cylindrical lens onto a first dye cell (dye concentration: 1 g/L). A spectrally narrow (10-nm-wide) part of the emitted light is selected by a dispersion-compensated double-pass grating spectrometer. We designed the setup to permit tunability (by simply moving a slit) without tem-
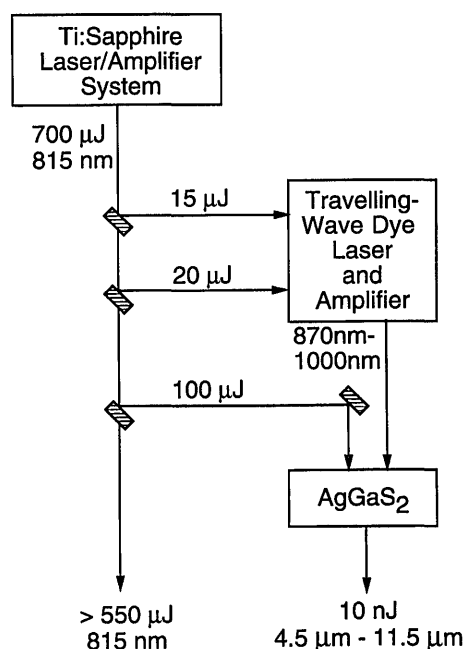

Fig. 1. Schematic of the experimental setup used to generate tunable subpicosecond pulses in the midinfrared.
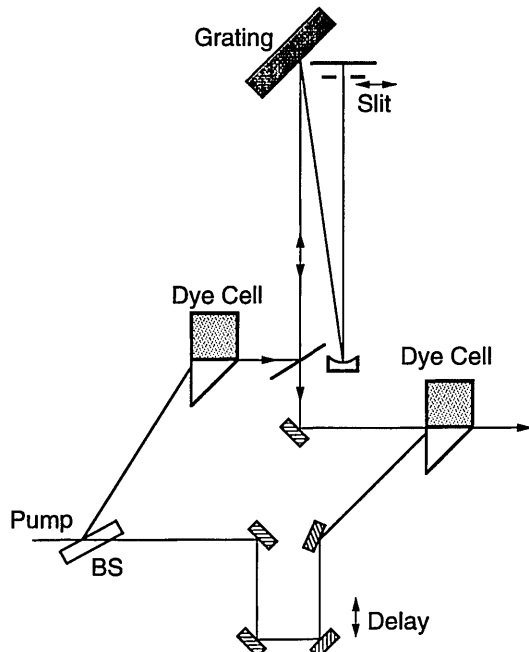
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Fig. 2. Schematic of the two-stage TWDL system. The output of a first TWDL cell is filtered in a grating spectrometer. The spectral portion that is selected by the slit is amplified in a second TWDL stage. BS, beam splitter.

poral dispersion. These seed pulses are amplified in a second TWDL stage that is pumped by approximately 20 μJ. More than 70% of the total energy from the TWDL amplifier is emitted at the wavelength of the seed pulse. With this configuration, a tuning range of 70 nm for a single laser dye can be achieved. This makes it possible to cover the whole wavelength region between 870 and 1000 nm by the use of just two laser dyes, namely, IR 140 (Lambda Physics) and IR 143 (Radiant Dyes).

A summary of the experimental data obtained for different near-infrared laser dyes is shown in Table 1. When only the first traveling-wave dye cell is used the duration of the light pulses is of the same order or even less than that of the pump pulses. Only for the dye IR 26, which emits further in the infrared at 1130 nm, are longer pulse durations found. With the two-stage setup, the duration of the tunable output pulses becomes longer. However, in the spectral range below 1000 nm the pulses are always shorter than 250 fs. The system permits an energy conversion of approximately 5%. A cross-correlation experiment was used to record the temporal jitter between the output of the TWDL system and the amplified Ti:sapphire pulses. Within the experimental

accuracy, no jitter is detected. This high degree of synchronization is essential for the generation of stable infrared pulses by difference-frequency mixing.

The nonlinear medium used in the difference-frequency generation process is a 1-mm-thick AgGaS$_2$ crystal cut with an angle of $\theta = 45°$ for critical collinear type I phase matching. Energies of $\approx$1 μJ from the tunable TWDL and 100 μJ from the Ti:sapphire amplifier with beam diameters of 1 mm are used. The energy of the midinfrared light is recorded by a calibrated HgCdTe detector after filtering with a Ge filter. This energy amounts to as much as 10 nJ, with fluctuations of less than 10%. The spectral properties of the midinfrared pulses are measured by a 25-cm grating spectrometer. Figure 3 shows several spectra of infrared pulses that we obtained by tuning the TWDL and rotating the phase-matching angle of the AgGaS$_2$ crystal. In this experiment we used only one laser dye (IR 140) in the TWDL system and obtained infrared pulses with center wavelengths between 6.5 and 11.5 μm and a spectral bandwidth of 60 cm^{-1}. With the dye IR 143 in the TWDL system, the tuning range extends to as low as 4.5 μm. The spectral bandwidth of the infrared pulses is limited by the phase-matching condition. The measured value of 60 cm^{-1} agrees well with theoretical considerations for a 1-mm-thick crystal.

The duration of the infrared light pulses was measured by means of a pump-and-probe experiment in which the change of the transmission through
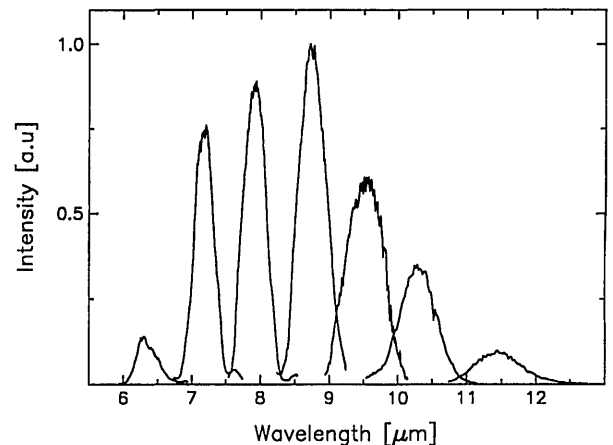

Fig. 3. Spectral properties of the midinfrared pulses for different wavelengths of the TWDL system and different phase-matching angles of the AgGaS$_2$ crystal. The central wavelength was tuned between 6.5 and 11.5 μm. Only one laser dye (IR 140) was used in this experiment.

Table 1. Properties of the Traveling-Wave Laser System

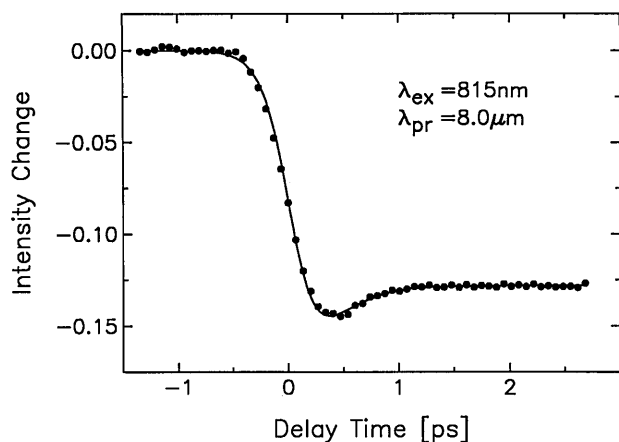| Laser Dye | Without Seed Pulse | | With Seed Pulse | | Total Energy Conversion Efficiency (%) |
	Central Wavelength (nm)	Pulse Duration (fs)	Tuning Range (nm)	Pulse Duration (fs)	
IR 140	875	150	870–930	150	5
IR 132	880	70	880–940	200	4
IR 143	920	95	920–1000	250	3
IR 26	1130	250	–	–	1

Fig. 4. Results of a pump-and-probe experiment on a thin Si plate. The intensity change of transmitted probing pulses (λ_{pr} = 8.0 μm) induced by the generation of hot carriers resulting from an intense near-infrared pulse (λ_{ex} = 815 nm) is plotted. A cross-correlation time of 450 fs can be deduced from the initial decrease of the signal intensity. The small change of the signal amplitude at longer delay times is presumably due to a fast cooling of the hot carriers.[6,13]

a silicon plate was detected (Fig. 4). A fraction of the near-infrared output of the Ti:sapphire laser illuminated the silicon plate, resulting in a fast decrease of the midinfrared signal that was due to the generation of hot carriers.[6] The changed transmission was monitored at a wavelength of 8 μm as a function of the delay time between the pulses. From modeling the initial decrease of the signal intensity, we can deduce a cross-correlation width of 450 fs. Deconvolution yields a duration of the infrared pulses of approximately 400 fs. Apparently the pulse duration of the infrared pulses is limited mainly by the group-velocity dispersion between the near-infrared pulses and the midinfrared pulses in the nonlinear crystal. A theoretical value of 300 fs is calculated for a 1-mm path length in the AgGaS$_2$ crystal. Therefore we expect to obtain even shorter pulses by use of a thinner crystal. From the measured pulse duration and spectral bandwidth, one calculates the bandwidth product to be $\Delta\nu\Delta t$ = 0.8, which is approximately two times the bandwidth limit for pulses with Gaussian pulse shape.

In the present setup the energy of the infrared pulses is limited by strong two-photon absorption of the intense 815-nm pump light in the AgGaS$_2$ crystal. This is because AgGaS$_2$ is a semiconductor with an absorption edge of approximately 470 nm. This determines the effective intensity that can be used for the downconversion process. Nevertheless,

it should be noted that no damage to the AgGaS$_2$ crystal is observed after several hundred hours of operation.

In a second setup we use the dye IR 26 in a one-stage TWDL in conjunction with a 3-mm-thick LiIO$_3$ crystal. This yields subpicosecond infrared pulses in the spectral range of 3 μm. Because of the absence of two-photon absorption in LiIO$_3$ at the pumping wavelength, higher infrared energies can be obtained. By use of only 100 nJ from the TWDL, the parametric light generation leads to midinfrared pulses with an energy of more than 200 nJ.

In conclusion, we have demonstrated a system that provides two independent and synchronized subpicosecond light sources in the midinfrared region suitable for subpicosecond pump-and-probe experiments. By mixing the output of a tunable TWDL system with the amplified Ti:sapphire pulses in a AgGaS$_2$ crystal, we can generate high-energy pulses between 4.5 and 11.5 μm. The pulse duration is 400 fs. In addition, in a simple setup in which IR 26 is used in a TWDL, intense subpicosecond pulses of approximately 3 μm can be obtained by use of a LiIO$_3$ crystal.

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