Infrared Pulses of 1 Picosecond Duration Tunable Between 4 μm and 18 μm

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Abstract—**Infrared pulses are generated via difference frequency mixing of Nd:glass laser pulses and infrared dye laser pulses. Tuning between 4 and 18 μm is achieved by various combinations of laser dyes (dye No. 5 and A 9860) and nonlinear crystals ($AgGaS_2$ and GaSe). The energy of the mid-infrared pulses is in the order of a μJ; the photon conversion efficiency is $\lesssim 2\%$. The duration of the nearly bandwidth limited pulses is measured to be 1 ps. The system operates with a repetition rate of 1 Hz.**

I. Introduction

THERE is considerable interest in tunable ultrashort laser pulses in the infrared. In the near infrared progress has been made by recently developed new laser materials. Tunable solid state lasers, e.g., color center, Ti:sapphire, Fosterite, and Co:MgF_2 lasers, and dye lasers, allow the generation of tunable pulses in selected regions between 0.7 μm and 2 μm.

The generation of widely tunable ultrashort pulses at longer wavelengths generally requires nonlinear optical processes. The process of lowest order is parametric three photon interaction which is based on the nonlinear susceptibility χ^2. Parametric amplification has been used for more than 20 years to generate tunable picosecond pulses in the UV, visible, and infrared spectral range [1]–[17]. For a long time single path systems [1]–[11] were preferred on account of their simplicity, but very recently parametric oscillators [12]–[17] have found increasing attention due to their capability of generating femtosecond pulses and due to their potential to be pumped by cw mode-locked lasers. At present time femtosecond pulses as short as 60 fs can be produced at special selected wavelengths [15], [17]. Broad tunability has been demonstrated for pulse durations of $\gtrsim 200$ fs and wavelengths $\lesssim 2$ μm. At wavelengths beyond 2 μm pulses of a few picoseconds have been generated by single path generators or by down conversion on crystals like $LiNbO_3$ [2]–[4], [8], $LiIO_3$ [18]. [19], BBO [18], proustite [20], and $AgGaS_2$ [7], [21], [22]. In particular, down conversion is of special interest due to superior properties of the generated tunable pulses.

In this paper the generation of tunable, ultrashort mid-infrared pulses by a parametric down conversion process is studied. A nonlinear crystal produces the difference frequency between the Nd:laser frequency and the tunable output of an infrared dye laser. The experimental system is similar to the set-up discussed in [21]. Here, substantial progress is reported: i) We start with a feedback controlled mode locked Nd:glass laser system which provides considerably shorter pump pulses of 2 ps duration. As a consequence, the temporal width of the mid infrared pulses is reduced to approximately 1 ps. ii) The tuning range is extended from 10 to 18 μm by introducing a different infrared laser dye and GaSe crystals.

II. Experimental

The experimental system is depicted in Fig. 1. A feedback controlled mode-locked (FCM) Nd:glass laser system [23] serves as a pump source for the parametric generator. The laser oscillator is adjusted for a pulse duration of 2 ps and high pulse to pulse stability. Amplification by three amplifier stages leads to pulse energies in the order of several mJ with a repetition rate of 1 Hz.

Laser pulses of approximately 1 mJ energy are sufficient to generate intense tunable mid-infrared pulses. A part of the pulses (~ 0.5 mJ) pumps a traveling wave infrared dye laser TWDL [24], which produces tunable pulses in the near infrared. A wavelength range from 1.1 μm to 1.4 μm is covered with the Q-switch dye A 9860 and the heptamethine pyrylium dye No. 5, respectively. Output energies up to 2 μJ are achieved in the near infrared. A second part of the Nd:glass laser pulses (~ 0.5 mJ) and the tunable output of the laser are mixed in a nonlinear $AgGaS_2$ or GaSe crystal. In this way, pulses at the difference frequency are generated with energies in the order of a μJ.

The pulse properties are characterized by several diagnostic systems. The energy is determined by a HgCdTe detector. An IR-spectrometer with various gratings measures the spectral position and the spectral width of the pulses. The pulse duration is obtained from cross correlation experiments between the IR pulses and the Nd:glass laser pulses in a thin $LiIO_3$ crystal.

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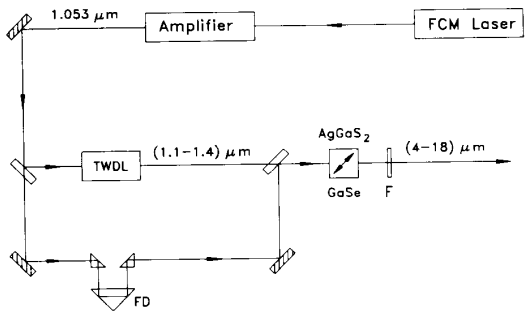

Fig. 1. Experimental system for the generation of tunable infrared picosecond pulses. The difference frequency between the FCM (feedback controlled mode-locked) Nd:glass laser and the tunable infrared TWDL (traveling wave dye laser) is generated in a $AgGaS_2$ or GaSe crystal. (FD = fixed delay, F = filter).

III. Nonlinear Crystals

Several parameters have to be considered in selecting the most suitable nonlinear crystal: i) The material should be highly transparent both at the pump and at the signal and idler wavelengths. ii) The nonlinear coefficients d_{eff} should be as high as possible. iii) The dispersion of the material has to allow phase matched difference frequency mixing. iv) The damage threshold has to be high enough.

There exist two materials with favorable properties for the parametric generation of mid-infrared pulses starting with a pump wavelength of 1.053 μm: $AgGaS_2$, which is commercially available, and GaSe. (Our GaSe crystals have been grown in the Azerb. Academy of Science.) Important parameters of the crystals are presented in Table I. $AgGaS_2$ limits the tuning range to wavelengths shorter than 12 μm due to its transparency range [25]. GaSe exhibits an absorption edge at 18 μm [10], due to phonon bands allowing the generation of infrared pulses up to this wavelengths.

The figure of merit d_{eff}^2/n^3 (n is the refractive index) given in Table I is larger by a factor of 600 for $AgGaS_2$ [30] and a factor 6000 for GaSe [26], [27] compared to KDP, respectively. Optical damage of the crystals is not observed for pump intensities below 1 GW/cm^2 after several months of operation.

Of special interest is the material GaSe which has been successfully used for difference frequency mixing of nanosecond pulses several years ago [28], [29] and of subnanosecond pulses quite recently [10]. In this paper, GaSe is introduced for the generation of tunable picosecond IR pulses. The negative uniaxial crystal belongs to the point group $\bar{6}2m$ and is grown by the Bridgman-Stockbarger method [26]. At present time it is not possible to polish GaSe under arbitrary angles. However, optically perfect surfaces are obtained by cleavage along (001) planes (z-cut). The high birefringence of the material (e.g., $n_o(1\ \mu m) = 2.91$ and $n_e(1\ \mu m) = 2.57$ [26]) results in small internal phase matching angles θ in our difference frequency generator. The angles θ for idler wavelengths $\lambda > 5\ \mu m$ are small enough to be reached with the z-cut.

TABLE I
Important Parameters of the Nonlinear Crystals $AgGaS_2$ and GaSe

	Transparency Range [μm]	Nonlinearity d_{eff} [10^{-11} m/V]	Figure of Merit d^2/n^3 (10^{-24} m^2/V^2
$AgGaS_2$	0.50-12	2^{30}	30
GaSe	0.64-18	8^{26}	300

The effective nonlinear coefficients depend on the phase matching and azimuthal (ϕ) angles in the following way [26]:

$$\text{type I } (e \rightarrow o + o)\text{: } d_{eff} = -d_{22} \cos\theta \sin 3\phi$$

$$\text{type II } (e \rightarrow o + e)\text{: } d_{eff} = -d_{22} \cos^2\theta \sin 3\phi$$

We use type I phase matching in our experiments requiring smaller angles θ. The highest effective nonlinearity is achieved by selecting azimuthal angles ϕ determined by $|\sin 3\phi| = 1$. There exist 6 orientations with maximum efficiency according to the symmetry of the crystal.

IV. Experimental Results

First we want to discuss the tuning curves of the nonlinear crystals. The data for $AgGaS_2$ can be found in [21]. For GaSe Fig. 2 shows the phase matching angle inside the crystal as a function of the idler wavelength for difference frequency with a pump wavelength of $\lambda = 1.053$ μm. The points represent the experimental results, the solid line is calculated from refractive index data in [26]. We observe good agreement between the experimental data and the calculated curve. The internal angles are located between 12° and 16° requiring an angle of incidence between 35° and 50° in z-cut crystals. The large angles result in considerable reflection losses. Nevertheless, the high nonlinearity of material allows an efficient generation of infrared pulses.

Before presenting the properties of the mid IR pulses comments should be made concerning the traveling wave infrared dye laser which provides input pulses at the signal frequency $\tilde{\nu}_s$ for the difference frequency mixing process. The dye laser system operates with Q-switch dyes characterized by fluorescence quantum efficiencies in the order of 10^{-3}. Fig. 3(a) shows the output energy as a function of the frequency. The frequency scale is chosen in such a way that the dye laser frequencies $\tilde{\nu}_s$ are plotted at the same abscissa positions as the corresponding idler frequencies $\tilde{\nu}_i = \tilde{\nu}_p - \tilde{\nu}_s$ in Fig 3(b) generated in the subsequent parametric three photon process ($\tilde{\nu}_p = 9497$ cm^{-1} is the Nd:glass laser frequency).

Dye No. 5 successfully used in TWDLs for several years gives a tuning range from 1.16 μm to 1.4 μm. A detailed discussion of the pulse properties is presented in [24]. Difference frequency mixing with these pulses result in a tuning range from 4 μm to 12 μm. For longer idler wavelengths another dye is required which supplies shorter signal wavelengths. Dye A 9860, for example, exhibits a blue shifted electronic transition compared to dye No. 5 [31]. Using dye A 9860 the tuning range of the

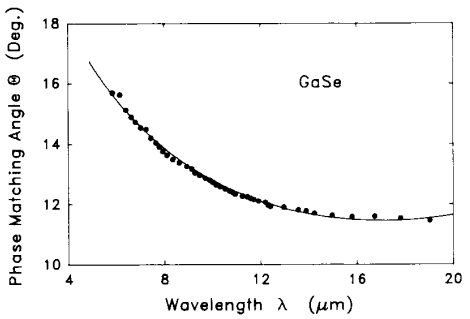

Fig. 2. Phase matching angle (angle between the optical axis and the k-vectors inside the crystal) as a function of the idler wavelength in GaSe. Experimental points are compared with a curve calculated using refractive index values taken from [26].

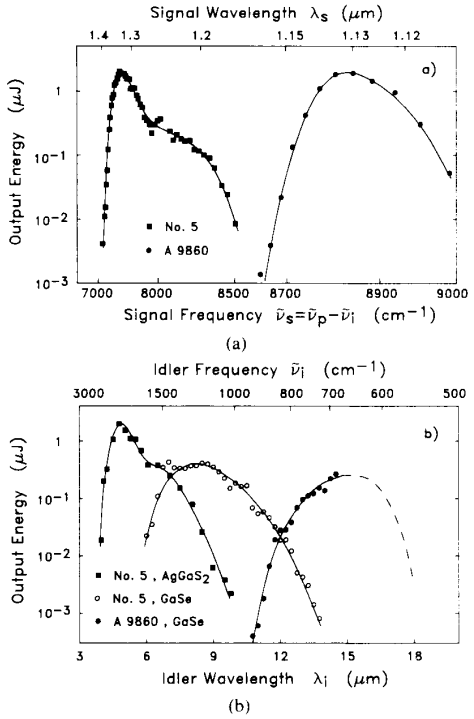

Fig. 3. (a) Output energy of the traveling wave infrared dye laser as a function of the frequency. Data for the dyes No. 5 and A 9860 are shown. The dye laser pulses serve as signal pulses in the parametric generator. The frequency scale is chosen in such a way that the (signal) frequencies are plotted at the same abscissa position as the corresponding idler frequencies in the lower part of the Figure. (b) Output energy of the idler pulses produced by the parametric difference frequency process as a function of the wavelength for three combinations of the laser dye and the nonlinear crystal.

TWDL shifts to wavelengths between 1.10 μm and 1.16 μm (see Fig. 3(a)) without changing the pulse duration and spectral width substantially. The blue shifted dye emission allows difference frequency generation at wavelengths up to 18 μm.

Most important is the output energy of the mid-infrared pulses achieved in our system. Fig 3(b) presents the pulse energy as a function of the wavelength for several combinations of the dye in the traveling wave laser and the nonlinear crystal. The energy of the infrared pulses at the maximum in Fig. 3(b) is in the order of a few μJ. The data are measured for crystals of 1 cm length. Fig. 3 shows that dye no. 5 has to be used for an efficient generation of pulses at wavelengths below 12 μm. For pulses at longer wavelengths dye A 9860 leads to higher output energies. Difference frequency mixing in $AgGaS_2$ is limited to wavelengths shorter than 10 μm to the absorption edge. A maximum photon conversion efficiency of approximately 2% is found at a wavelength of 5 μm. Higher conversion efficiencies may be achieved with an improved quality of the beam profile.

In the combination dye No. 5 and a $AgGaS_2$ crystal the energy of the mid-infrared pulse is strongly determined by the dye laser output energy at the corresponding signal frequency. The wavelengths dependence in Fig. 3(b) closely follows the curve for dye No. 5 in Fig. 3(a). The energy of the idler pulses decreases with increasing wavelength. For wavelengths longer than 7 μm GaSe of 1 cm length gives a higher output energy due to the higher nonlinearity of the material.

Replacing $AgGaS_2$ by GaSe a difference frequency dependence is observed in Fig. 3(b). The conversion efficiency is reduced with decreasing wavelengths for $\lambda < 8$ μm due to a rise of the angle of incidence of the GaSe crystal connected with growing losses due to the limited aperture and due to the increasing reflection of the z-cut crystal. At longer wavelengths the higher nonlinearity of GaSe results in an output energy exceeding that of $AgGaS_2$ by more than a factor of ten. A tuning range up to 13 μm is observed.

For wavelengths beyond 12 μm the combination dye A9860 and GaSe is superior in output energy (see Fig. 3(b)). Pulses of sufficient energy at the corresponding signal wavelengths shorter than 1.15 μm can be supplied only by dye A 9860. Comparison of the dye laser emission in Fig. 3(a) and the wavelength dependence of the mid-infrared pulses in Fig. 3(b) shows a similarity of the curves referring to a strong dependence of the idler energy on the dye laser output energy. In Fig. 3(b) experimental points are shown only for wavelengths shorter than 15 μm limited by the sensitivity range of our HgCdTe detector. The broken line is determined indirectly via the amplification of the corresponding signal pulses. Amplification down to a wavelength of $\lambda = 1.118$ μm is observed corresponding to an idler wavelength of 18 μm close to the absorption edge of the crystal. Typical amplification factors are between 10 and 100.

The energy of the idler pulses as a function of the pump intensity I_p is shown in Fig. 4 for the frequency $\tilde{\nu}_i = 1210$ cm^{-1}. Experimental data for three lengths of the GaSe crystal, $l = 1.7$ mm, $l = 4$ mm, and $l = 10$ mm, are plotted. We observed a rapid rise of the energy as a function of the pump intensity over several orders of magnitude. At pump intensities beyond 100 MW/cm^2 saturation of the parametric amplification process is clearly

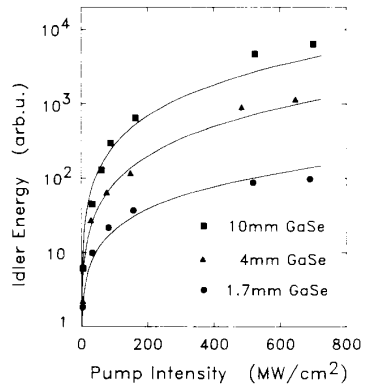

Fig. 4. Dependence of the idler energy on the pump intensity for a GaSe crystal of 1.7 mm, 4 mm, and 10 mm length, respectively. A rapid rise of the idler energy is found. At pump intensities beyond 100 MW/cm^2 saturation is clearly observed. As expected from theory the parametrically generated idler energy scales with $I_p l^2$ within the experimental error.

observed. The difference frequency generator is operated in the high gain limit [32] ($\Gamma l > 1$, where $\Gamma \propto \sqrt{I_p}$ is the parametric gain coefficient).

In this case, the generation of an idler pulse via down conversion is followed by exponential parametric amplification of both the signal and idler pulses according to exp ($2\Gamma l$). Finally, saturation occurs due to the depletion of the pump pulse. Amplification factors up to 100 are measured for the signal pulse. At pump intensities above 800 MW/cm^2 a more gradual rise of the output energy connected with rising pulse durations is expected. Comparing the three curves in Fig. 4 we find approximately the same idler energy for the same value of $\sqrt{I_p l}$ which is expected for crystals of the same quality [32].

Next, we want to discuss the spectral properties of the mid infrared pulses. They are strongly determined by the bandwidths of the incoming traveling wave dye laser pulses [21], which do not depend on the frequency and which are determined by the properties of the spectral selection unit [24]. Only minor contributions to the spectral widths are expected from the parametric amplification process on account of the dispersion of the crystals [5]. Spectral broadening on account of the pump beam divergence, which is smaller than 1 mrad, is estimated to be negligible. Consequently, the bandwidths of the mid-infrared pulses are nearly independent of the frequency position. Fig. 5 shows the spectral intensity of idler pulses at two frequency positions a) $\tilde{\nu}_i = 988$ cm^{-1} ($\lambda = 10.1$ μm) and b) $\tilde{\nu}_i = 1395$ cm^{-1} ($\lambda = 7.2$ μm). The data are taken with dye No. 5 in the traveling wave dye laser and a GaSe mixing crystal of 4 mm length. A spectral width of approximately 12 cm^{-1} and 15 cm^{-1} is observed for the idler frequencies $\tilde{\nu}_i = 988$ cm^{-1} and $\tilde{\nu}_i = 1395$ cm^{-1}, respectively (Fig. 5(a) and 5(b)).

Of special interest is the duration of the mid-infrared pulses. The temporal characteristics are investigated by cross correlation measurements between the mid-infrared pulses and Nd : glass laser pulses. The sum frequency be-

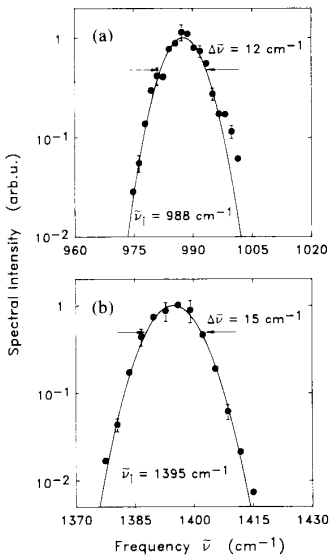

Fig. 5. Spectral intensity distribution of idler pulses generated with a 4 mm GaSe crystal. Experimental data for two frequency positions (a) of $\tilde{\nu}_i = 988$ cm^{-1} and (b) $\tilde{\nu}_i = 1395$ cm^{-1} are shown. The spectral widths of 12 cm^{-1} and 15 cm^{-1} are determined by the spectral properties of the dye laser pulses and by the dispersion of the nonlinear crystal.

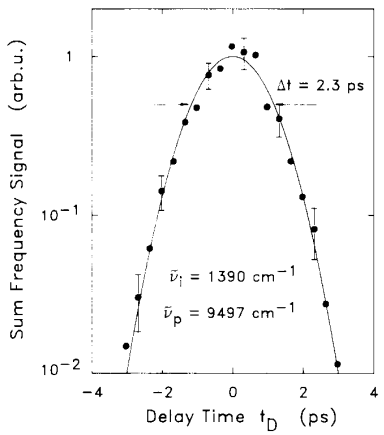

Fig. 6. Cross correlation curve of an idler pulse at $\tilde{\nu}_i = 1390$ cm^{-1} generated with a 4 mm GaSe crystal and the fundamental pulse of the Nd : glass laser at $\tilde{\nu}_p = 9497$ cm^{-1}. The solid line is calculated for Gaussian shaped pulses and a cross correlation width of $\Delta t = 2.3$ ps. Deconvolution with a duration of 2.1 ps for the Nd : glass laser pulse gives a duration of 1 ps for the idler pulse.

tween the two pulses is generated in a $LiIO_3$ crystal of 1 mm length. Fig. 6 shows the experimental result for pulses at $\tilde{\nu}_i = 1390$ cm^{-1} ($\lambda = 7.2$ μm) generated by a 4 mm GaSe crystal. The solid line is calculated for Gaussian shaped pulses and for a halfwidth of the cross correlation curve of 2.3 ps. The curve fits the experimental data over two orders of magnitude with high accuracy. The duration of Nd : glass laser pulses was determined in a separate experiment to be 2.1 ps. Deconvolution of the data in Fig. 6 gives a duration of the pulses in the mid-infrared of 1

ps. The relatively low group velocity dispersion of $1/v_g^p - 1/v_g^i = 1$ ps/cm allows the observed pulse shortening due to gain narrowing [13]. With a GaSe mixing crystal of 1 cm length a slightly longer pulse duration of approximately 1.9 ps is measured.

The pulse duration determined in Fig. 6 together with the spectral width in Fig. 5(b) results in a bandwidth product of 0.5 for pulses at $\lambda = 7.2$ μm close to the theoretical value for Gaussian pulses. At longer wavelengths in the range of $\lambda = 10$ μm we expect a slightly longer pulse duration due the reduced spectral width in Fig. 5(a) and due to an increase in group velocity dispersion by approximately 50%. A direct determination of the pulse duration is not possible due to the absorption of our correlation crystal at these wavelengths.

V. Summary

A laser system is presented which generates tunable pulses between 4 μm and 18 μm. The output energy depends on the generated wavelength; it is in the range of several 10 nJ up to a few μJ. The intensity of the pulses amounts to several 100 MW/cm^2. The divergence is determined by the divergence of the traveling wave dye laser [24]; it is in the order of a few mrad allowing efficient focusing of the pulses. Pulse durations down to 1 ps are observed. The extension of the tuning range to longer wavelength is a result of the introduction of the highly efficient GaSe crystals which exhibit a transparency range far into the infrared.

At present time the mid-infrared pulses are applied to study intersubband scattering in quantum well structures by a bleaching technique. The intensity of the pulses allows efficient excitations and the pulse duration is short enough to investigate the relevant time constants. The results are discussed elsewhere.

Finally, we want to point out that down conversion in $AgGaS_2$ and GaSe works also with other pump wavelengths. Generally, the absorption determines the shortest pump wavelength which can be used, but in both crystals decreasing pump wavelengths restrict the tuning range to longer idler wavelengths. In $AgGaS_2$ the tuning range is given by the phase matching properties [33]. In GaSe phase matching is generally possible for $\lambda_p > 0.7$ μm [26]. For $\lambda_p < 1.5$ μm, however, the relevant angles for wavelengths close to the degeneracy point are too large to be reached with z-cut crystals.

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