Ultrafast Mid-Infrared Pulse Generation

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Received March 20, 1995

We discuss the applications of short mid-infrared pulses for time resolved spectroscopic measurements in several different areas. A review of the techniques used to generate mid-infrared pulses is presented. Recent results are described on mid-infrared pulse generation using a two color mode- locked Ti:sapphire laser. The infrared pulses are tuned from 7.5 to 12.5 μm and have duration of 500 fs.

I. Introduction

Most of the early investigation of ultrafast phenomena was restricted to the visible spectral region, due to the limited number of available laser sources. Recently there has been a progressive increase in the spectral range of the short pulses generated. In particular, experimental systems based on ultrashort mid-infrared (mid-IR) pulses have proved to be powerful tools to investigate fundamental processes in many different materials.

Mid-IR pulses have been used to study carrier dynamics in semiconductors. Experiments of visible pump and mid-IR probe have been used to study photogenerated carrier dynamics in semiconductors. Mid-IR pulses can probe the carriers far from the surface, since most of the semiconductors are transparent to mid-infrared radiation. Additionally, information on the spatial profile of the carriers can be obtained from the infrared reflectivity[1].

Picosecond pulses at 4.83 μm were used to study the dynamics of photogenerated carriers in InP:Fe[2]. Intervalley scattering dynamics in GaAs have also been studied by using UV pump and mid-IR probe[3]. Picosecond pulses with wavelengths ranging from 5 to 10 ,um were utilized to measure free carrier absorption coefficient in InAs and GaAs[4]. This experiment allowed to investigate hot LO-phonon thermalization in polar semiconductors. Mid-IR pulses are essential tools to investigate intervalence band transitions in semiconductors. Femtosecond pulses from 2.7 to 5.0 μm were used to perform measurements of hole dynamics in bulk Ge[5]. An experiment of mid-IR pump and visible probe was used to investigate hole capture by shallow acceptors in p-type GaAs[6]. Still in bulk semiconductors, mid-IR pulses were used in pump and probe experiments in narrow gap semiconductors, such as PbSe[7] and $Hg_{1-x}Cd_xTe$[8].

In quantum wells, intersubband transitions were investigated using ultrafast mid-IR pulses[9-10]. The transitions between two bound states in a quantum well were first observed in the conduction band[11] and, more recently, in the valence band[12]. The intersubband transition times can be measured by visible pump and mid-IR probe[10] or both pump and probe in the mid-IR wavelengths[9], being the choice of the pump wavelength related to the doping level of the samples.

Mid-infrared pulses are required in some technological applications, such as, to characterize optoelectronic devices designed for these frequencies. The pulses were used to measure the response time of fast infrared detectors based on intersubband transitions in quantum wells[13,14].

In photochemistry, multiphoton spectroscopy is

only possible due the existence of mid-IR radiation, since this process requires the absorption of many photons (typically 10- 40)[15]. There are many questions related to the molecular kinetics during a multiphoton excitation that can be addressed using ultrashort mid-IR pulses.

Another application of mid-IR pulses is to investigate vibrational states of molecules adsorbed on surfaces. Visible pump and mid-IR probe were used to study surface vibrational energy relaxation of CO on Cu[16].

Mid-IR spectroscopy has many applications in biochemistry. Ultrafast pulses were used to study dynamics of ligand motion in hemoglobin[17,18] and in myoglobin[19] by measuring vibrational relaxation of the CO or O_2 molecules.

II. Techniques for ultrashort mid-infrared pulse generation

Several different techniques have been used to obtain mid-infrared pulses. These techniques include color center lasers, optical parametric oscillators, semiconductor switching, and difference frequency mixing.

Passive mode-locking in color center lasers have allowed to produce short pulses at wavelengths up to 2.9 μm [20-22] and optical parametric oscillators (OPO), pulses at wavelengths up to 3.65 μm. [23 24] Both techniques provide high average power and high repetition rate pulses.

Mid-infrared pulses have also been generated by semiconductor switching[25-27]. In this technique, a semiconductor is illuminated simultaneously by amplified visible pulses and CO_2 laser pulses at 10.6 μm. The high power visible pulses create a dense free carrier plasma that reflects the mid-infrared light. A combination of two CdTe switches allowed to generate 130 fs pulses at 100 HZ.[26] Recently 1 ps pulses were obtained using a single GaAs switch[27].

The difference frequency mixing approach requires sources of two frequencies. This can be obtained with several systems, such as a high power amplifier based system, two synchronously mode-locked lasers, a CW and a pulsed laser, or an optical parametric oscillator. In the amplifier based system, the pulses are used to generate a continuum, and difference frequency mixing is then performed between the amplified pulses from the laser and a selected range of the continuum[28,29]. A similar technique involves the use of a traveling wave dye cell[30,31]. With amplifier based systems one can obtain tunable infrared pulses with high pulse energies and repetition rates in the range of several kHz.

Another technique consists in mixing the pulses from two separate lasers, being one syncronously pumped by the second harmonic of the other[32]. This has yielded picosecond pulses tunable from 3.4 to 7.0 μm at a repetition rate of 76 MHz with an average power of 15 μW. It is also possible to mix short pulses with light from a CW laser. Pulses at 2.6 to 4 μm were obtained by mixing the frequencies from a Nd:YAG and a color center lasers, in a KNbO3 crystal[33]. The pulse duration was 6 ps and the average power was 1.5 mW.

The last technique consists in mixing the signal and the idler pulses from a Ti:Sapphire pumped OPO cavity. The mid-infrared pulses generated using this technique have wavelength ranging from 2.6 to 5.0 ?m and a repetition rate of 80 MHz[34].

The propagation of mid-IR pulses through atmospheric air causes distortions due to absorption by CO_2 and H_2O molecules. Even if the detuning from the resonances is large enough to prevent the beam energy from being absorved, the dispersion due to these resonances can seriously distort the pulses[35]. The absorption spectrum of 40 cm of air is shown in Fig. 1. In the range from 2.5 to 13 μm, atmospheric air has three sets of absorption lines. H_2O molecules absorb around 2.7 μm and from 5 to 7.5 μm. The line around 4.25 μm is due to CO_2 absorption. In order to avoid these distortions, one has to place the mid-IR system in a flow box filled with dry nitrogen.

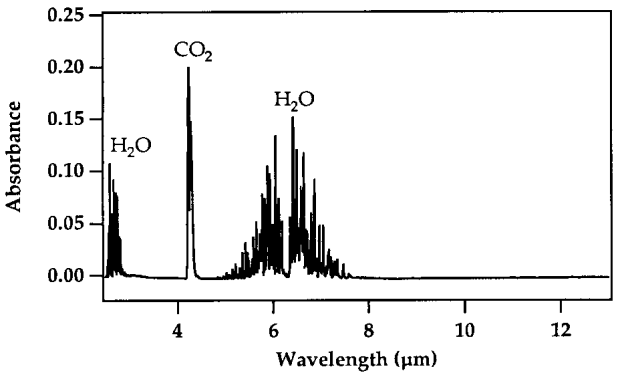

Figure 1. Absorption spectrum of air in the wavelength range from 2.5 to 13 μm.

III. Mid-infrared pulses generated from a two color Ti:sapphire laser

Ultrafast mid-IR pulses are generated by difference

frequency mixing of the frequencies from a two color femtosecond mode-locked Ti:sapphire laser[36].

The two color Ti:sapphire laser is tuned to generate synchronous pulses with center wavelengths of 780 and 840 nm. The pulse durations are 60 fs. The experimental setup used to perform the difference frequency mixing between the collinear pulses from the two color laser is showed in Fig. 2. The setup allows for independent compensation of the group velocity dispersion at the two wavelengths, as well as for the adjustment of the temporal overlap between the two pulses.

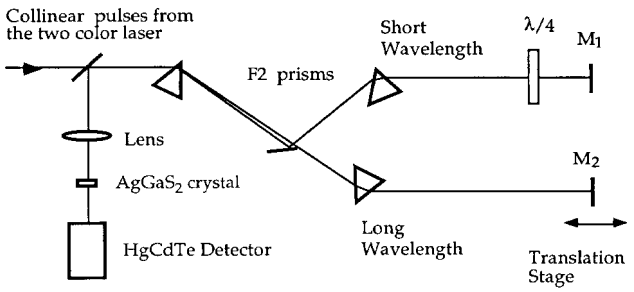

Figure 2. Experimental system used to generate mid-IR pulses from a two color Ti:sapphire laser.

Both beams are focused on a 1.0 mm thick $AgGaS_2$ crystal cut at 39° with respect to the optical axis. The mixing is performed using type I phase-matching ($e = o + o$, for $\omega_{SW} = \omega_{LW} + \omega_{IR}$). The average power of each of the near infrared beams, just before the mixing crystal, is 60 mW. The average power obtained at a wavelength of 10 μm is 5 μW, measured using a liquid nitrogen cooled HgCdTe detector. A germanium filter is used to block the near infrared beam. The repetition rate of the mid-IR pulses is 82 MHz.

The tuning curve obtained by rotating the $AgGaS_2$ crystal, for a fixed separation of 60 nm between the center wavelengths of the near-infrared pulses, is shown in Fig. 3. The peak wavelength corresponds to the difference between the center frequencies of the two pulse trains from the Ti:sapphire laser. It is also possible to increase the tunability of the mid-IR pulses by tuning the separation between the two wavelengths from the laser.

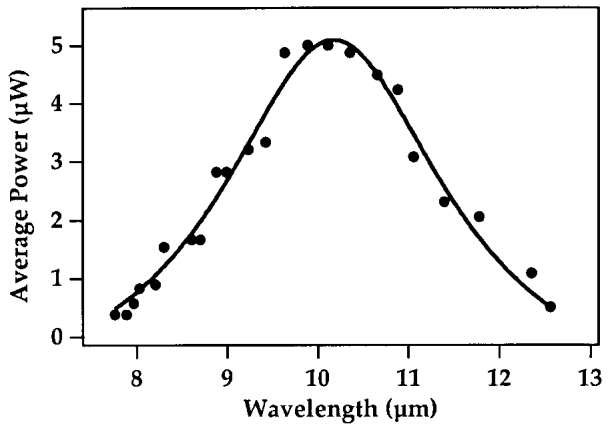

Figure 3. Tuning curve of the mid-IR pulses, obtained by tuning the $AgGaS_2$ crystal.

The tuning range of the center wavelength of the mid-IR pulses is 7.5 to 12.5 μm (165 to 99 meV or, in wavenumbers, 1,333 to 800 cm^{-1}), as the angle between the input beam and the normal to the crystal surface is varied from 25° to 3°. This corresponds to internal angles between 49° and 40.2°, as shown in Fig. 4. In this figure, the dots represent experimental data and the smooth line was calculated using Sellmeia equations.

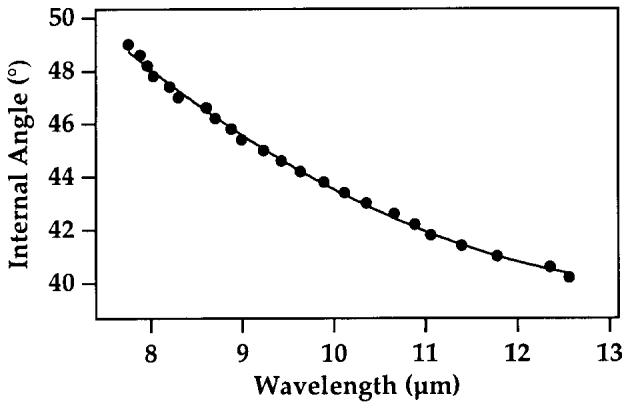

Figure 4. $AgGaS_2$ crystal internal angle versus wavelengths obtained in the mid-IR generation.

IV. Pulsewidth measurement

The pulsewidth of the mid-IR pulse is obtained by a cross-correlation of the mid-IR pulse and the pulse centered at 780 nm from the two color laser. For this experiment, the mirror M1, shown in Fig. 2, was replaced by a 50% beam splitter. The transmitted fraction of the 780 nm beam traverses a delay line and is mixed with the mid-IR pulses in a second $AgGaS_2$ crystal. This crystal is cut at 45° and is 1.0 mm thick. Collinear configuration is used. The upconverted light is detected

using a GaAs cathode photo- multiplier tube and the signal is integrated using a lock-in amplifier. The upconverted pulses have a center wavelength near 730 nm. Stray light from the 780 nm beam is rejected from the detector by means of two interference filters centered at 730 nm. The beam at 780 nm is also filtered with a low pass filter, immediately prior to the upconversion crystal, so as to eliminate any radiation centered at 730 nm coming directly from the laser. The upconversion is performed using type I phase-matching.

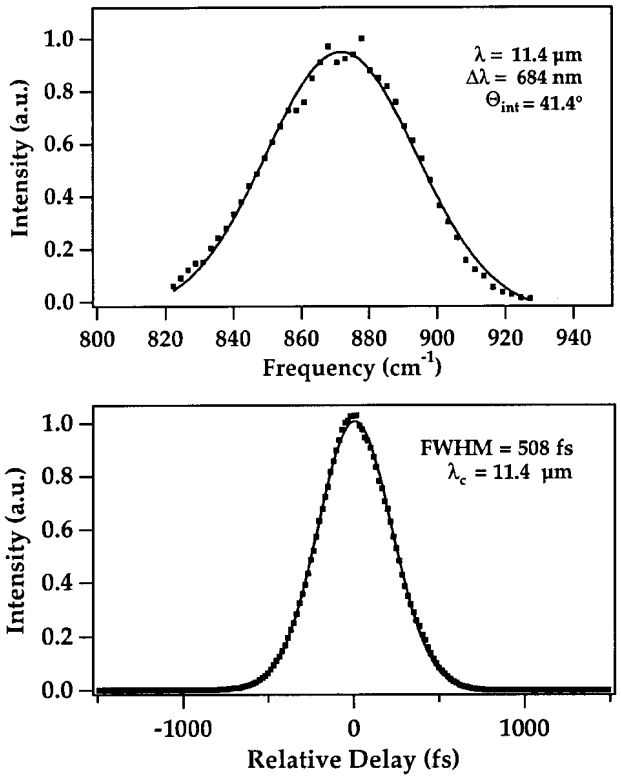

Figure 5. Spectrum and temporal profile of a pulse centered at the wavelength of 11.4μm. The spectral width is 684 nm and the pulse duration is 500 fs.

The cross-correlation profile obtained for $\lambda = 11.4$ μm is shown in Fig. 5. The full width at half maximum of the cross correlation curve is 508 fs. Before the sum frequency mixing, the pulse at 780 nm traverses a beam splitter, a quarter-wave-plate, a low pass filter and a lens. Due to dispersion introduced by these elements, the pulse at 780 nm has a duration of 95 fs. Deconvolving the 95 fs duration of the near infrared pulses from the cross-correlation width yields a full width at half maximum of 500 fs for the intensity profile of the mid-IR pulses. The time-bandwidth product $\Delta t \Delta \nu$ is near to 0.7, which corresponds to 1.6 the transform limit for Gaussian pulses.

The finite thickness of the nonlinear crystal limits the minimum pulse duration achievable in the difference frequency mixing process. This happens because there is a temporal walk off between the near infrared and mid infrared pulses, as they traverse the $AgGaS_2$ crystal. This walk off arises from the fact that the near infrared pulses and the mid-infrared pulse travel at different speeds in the material medium. For instance, the group velocity mismatch between the wavelengths of 840 nm and 10μm, through 1 mm of $AgGaS_2$, is 400 fs. This effect prevents one from obtaining transform limited pulses, since the infrared radiation is going to be generated over a wide temporal range. Another reason why the pulse duration depends on the crystal thickness is related to the fact that the phase matching condition is valid for a limited range of the infrared frequencies. This means that, for a given crystal thickness and angle, only a limited width of spectrum is in phase matching condition. This limitation has the same effect as a filter on the mid-infrared generation.

In conclusion, we presented a review of the applications of mid-IR pulses and the techniques used to generated ultrafast mid-IR pulses. We have reported the generation of wavelength tunable mid-infrared pulses by difference frequency mixing between pulses from a single cavity laser source. This is the first source of femtosecond pulses at high repetition rate in the 7.5 to 12.5 μm range. Higher peak powers can be obtained by amplifying the two color pulses from the Ti:sapphire laser in a multipass amplifier[37,38], before doing the difference frequency mixing.

We would like to acknowledge M. M. Broer, L. Rothberg, P. R. Smith, and J. D. Kafka for helpful discussions. M. R. X. de Barros thanks the Brazilian agency Conselho Nacional de Desenvolvimento Científico e Tecnológico, for financial support.

References

1. R. S. Miranda, H. W. K. Tom, A. M. Johnson, T. J. Bridges, and G. D. Aumiller, Opt. Lett. **16**, 1859 (1991).
2. R. S. Miranda, H. W. K. Tom, A. M. Johnson, T. J. Bridges, and G. D. Aumiller, Appl. Phys. Lett. **60**, 1105 (1992).

3. M. A. Cavicchia, R. R. Alfano, Ultrafast Phenomena IX, paper MD29, pg. 128 (1994).
4. T. Elsaesser, R. J. Bauerle, R. Klann, and W. Kaiser, Ultrafast Phenomena VII, pg. 328 (1990).
5. M. Woerner, W. Frey, M. T. Portella, C. Ludwig, T. Elsaesser, and W. Kaiser, Phys. Rev. B **49**, 17007 (1994).
6. A. Lohner, M. Woerner, T. Elsaesser, and W. Kaiser, Ultrafast Phenomena VIII, pg. 416 (1992).
7. C. Ludwig, W. Frey, M. Woerner, T. Elsaesser, Opt. Commun. **102**, 447 (1993).
8. A. V. Nurmikko and B. D. Schwartz, J. Vac. Sci. Technol. **21**, 229, (1982).
9. A. Sellmeier, H. J. Hübner, G. Abstreiter, G. Weimann, W. Schlapp, Phys. Rev. Lett. **59**, 1345 (1987).
10. R. J. Bauerle, T. Elsaesser, H. Lobentanzer, W. Stolz, K. Ploog, Phys. Rev. B **40**, 10002 (1989).
11. L. C. West, S. J. Eglash, Appl. Phys. Lett. **46**, 1156 (1985).
12. J. S. Park, R. P. G. Karunasiri, K. L. Wang, Appl. Phys. Lett. **61**, 681 (1992).
13. C. G. Bethea, B. F. Levine, G. Hasnain, J. Walker, R. J. Malik, J. Appl. Phys. **66**, 963 (1989).
14. M. R. X. de Barros, P. C. Becker, R. S. Miranda, L. C. West, J. Dunkel, C.W.Roberts, J. W. Stayt, Jr. and V. Swaminathan, Electron. Lett. **30**, 1093 (1994).
15. M. Quack, Infrared Phys. **29**, 441 (1989).
16. S. F. Shane, L. Rothberg, L. H. Dubois, N. J. Levinos, M. Morin, and A.L. Harris, Ultrafast Phenomena VII, pg. 362 (1990).
17. T. Jedju, L. Rothberg, A. Labrie, Opt. Lett. **13**, 961 (1988).
18. R. M. Hochstrasser, P. A. Anfinrud, R. Diller, C. Han, M. Iannone, T. Lian, and B. Locke, Ultrafast Phenomena VII, pg. 429 (1990).
19. M. Lin, T. A. Jackson, P. A. Anfinrud, Ultrafast Phenomena IX, paper FB4, pg. 597 (1994).
20. C. L. Cesar, M. N. Islam, C. E. Soccolich, R. D. Feldman, R. F. Austin, and K. R. German, Optics. Lett. **15**, 1147 (1990).
21. P. N. Kean, X. Zhu, D. W. Crust, R. S. Grant, N. Langford, and W. Sibbet, Opt. Lett. **14**, 39 (1989).
22. C. L. Cesar, M. N. Islam, C. E. Soccolich, R. D. Feldman, R. F. Austin, and K. R. German, Ultrafast Phenomena VII, pg. 17 (1990).
23. P. E. Powers, C. Tang, L. K. Cheng, Opt.Lett., **19**, 1439 (1994).
24. R. A. Crowell, G. R. Holtom, S. X. Xie, Ultrafast Phenomena IX, Paper ThD23 (1994).
25. P. B. Corkum, Opt. Lett. **8**, 514 (1983).
26. C. Rolland and P. B. Corkum, J. Opt. Soc. Am. B**3**, 1625 (1986).
27. A. Y. Elazzabi, J. Meyer, M. K. Y. Hughes, and S. R. Johnson, Opt. Lett. **19**, 898 (1994).
28. P. C. Becker, D. Gershoni, Ultrafast Phenomena VII, (Springer Verlag, Berlin, 1990) p.81.
29. T. Jedju, L. Rothberg, Appl. Opt.**27**, 615 (1988).
30. T. Elsaesser, M. C. Nuss, Opt. Lett. **16**, 411 (1991).
31. P. Hamm, C. Lauterwasser, W. Zinth, Opt. Lett. **18**, 1943 (1993).
32. A. G. Yodh, H. W. K. Tom, G. D. Aumiller, R. S. Miranda, J. Opt. Soc. of Am. B**8**, 1663 (1991).
33. U.Socha, J. Hussong, R. Urschel, A. Nebel, R. Beigang, Conference on Laser and Electro Optics'94, paper CThQ2, pg 378 (1994).
34. J. D. Kafka, M. L. Watts, J. W. Pieterse, Conference on Laser and Electro-Optics, Vol. 8, OSA Technical Digest, paper CWI6 (1994); J. D. Kafka, M. L. Watts, J. W. Pieterse, Ultrafast Phenomena IX, Paper TuD21 (1994).
35. A Seilmeier, M. Worner, H.-J. Hbner, and W. Kaiser, Appl. Phys. Lett. **53**, 2468 (1988).
36. M. R. X. de Barros, P. C. Becker, Opt. Lett. **18**, 631 (1993).
37. C. LeBlanc, G. Grillon, J. P. Chambaret, A. Migus, and A. Antonetti, Opt. Lett. **18**, 140 (1993).
38. J. Zhou, C.-P. Huang, C. Shi, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **19**, 126 (1994).