Appl. Phys. B 64, 619–621 (1997)

Applied Physics B Lasers and Optics

Rapid communication

Coherent generation of tunable, narrow-band THz radiation by optical rectification of femtosecond pulse trains

C. Messner*, M. Sailer*, H. Kostner*, R.A. Höpfel

Institut für Materialphysik, Universität Wien and Ludwig-Boltzmann-Institut für Festkörperphysik, A-1090 Vienna, Austria
(Fax: +43-512/507-2921, E-mail: christina.messner@uibk.ac.at)

Received: 14 March 1997

Abstract. We report coherent generation of tunable THz pulses via optical rectification of pulse trains consisting of femtosecond laser pulses. The synthesis of pulse trains is performed with zone plates made of thin glass slides to achieve both tunability and a narrower spectral bandwidth of the generated THz radiation. For the first time, tuning of the far infrared radiation has been experimentally demonstrated from 2.0 to 5.3 THz.

PACS: 42.72 Ai; 42.79 Ci; 42.65Ky; 42.65 An

Over the last decade, continuous progress has been made in the generation of coherent subpicosecond ultrashort pulses of terahertz (THz) radiation in free space. Various techniques have been developed using high speed photoconductors as fast current sources for radiating antennas [1, 2] as well as coherent tunneling in quantum wells [3, 4] and Bloch oscillations in superlattices [5]. A different approach is the rectification of optical pulses in semiconductors [6, 7] and electro-optic crystals [8–11]. The tunability achieved in recent experiments covers a spectral range up to ∼2 THz [12–15]. Several applications such as THz time domain spectroscopy [16], THz imaging [17] and optical pump, THz-probe spectroscopy [18] have already been demonstrated. However, there is still a lack of a pulsed, tunable, and coherent radiation source in the spectral region between 2 and 10 THz (8 to 40 meV), which represents the energy range of many electronic, phononic and molecular excitations.

In this work we present a new and very powerful method for the generation of ultrashort, coherent far infrared (FIR) pulses. This method is based on the synthesis of THz *rate, optical pulse trains* by means of a zone plate of stacked glass slides. Optical rectification of these pulse trains in a material of large optical susceptibility $\chi^{(2)}$ yields narrow-band THz pulses. The THz frequency can easily be varied by tilting the zone plate. The simple construction of the zone plate allows easy control of all parameters of the generated pulse trains. Therefore frequency, envelope, and chirp of the THz pulses can be arbitrarily designed. Common pulse shapers [13] consist of a grating pair and a phase or amplitude mask in the Fourier plane which leads to significant intensity loss. On the other hand, in our system the energy of a single optical pulse is totally converted into the pulse train.

Using a zone plate of four glass slides we could experimentally demonstrate a substantial narrowing of the spectral bandwidth of the THz radiation. In addition, the frequency was tuned from 2.0 to 5.3 THz by using three zone plates with glass slides of different thicknesses and by tilting the zone plates. We also found excellent agreement of our experimental data with theoretical calculations.

Optical rectification [19] of femtosecond laser pulses in electro-optic crystals occurs due to the nonlinear optical susceptibility $\chi^{(2)}$ which gives rise to a second harmonic term and a transient DC polarisation $\boldsymbol{P}_0$ with the same time dependence as the intensity of the excitation laser pulse. The electric field of the emitted THz radiation $\boldsymbol{E}_{\mathrm{THz}}$ is proportional to the second time derivative of $\boldsymbol{P}_0$ and hence:

$$I_{\mathrm{THz}}(t) \propto |\boldsymbol{E}_{\mathrm{THz}}|^2 \propto \left|\frac{\partial^2 \boldsymbol{P}_0}{\partial t^2}\right|^2 \propto \left(\chi^{(2)} I_{\mathrm{optical}}(t)\right)^2 . \qquad (1)$$

A single optical pulse therefore generates a THz electric field of one and a half cycles. However, using optical pulse trains consisting of several pulses with a THz repetition rate, more cycles of $\boldsymbol{E}_{\mathrm{THz}}$ can be synthesized, which results in a narrowing of the spectrum. A quasi-sinusoidal electric field of the emitted THz pulse is obtained if the time delay between the pulses of an equally spaced, Gaussian shaped, pulse train is chosen to be 1.47 times the FWHM of a single optical pulse.

The experimental setup is shown in Fig. 1. The source of optical pulses was a mode-locked Ti:Sapphire laser at a repetition rate of 76 MHz with typical pulse energies of 15 nJ at a central wavelength of 850 nm. An external prism stage controlled the pulse width giving nearly transform limited pulses of 70 fs. Pulse trains were created from single optical pulses by means of zone plates consisting of four glass slides equally shifted with respect to each other (see inset in Fig. 1). Passing through a single slide with a refractive index n and thickness d, the optical pulse is delayed by $\Delta t = d(n_{\mathrm{g}} - 1)/c$ where c/n_{g}

*Experimental work performed at the University of Innsbruck, Austria

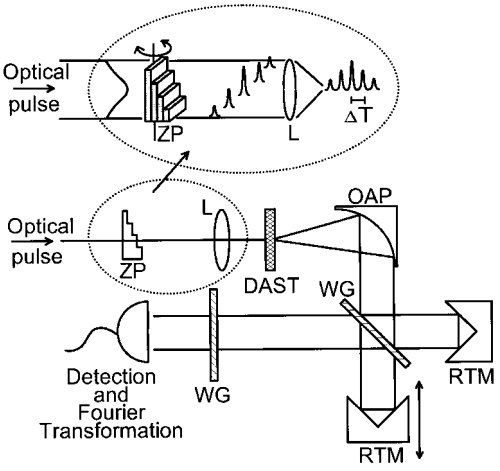

Fig. 1. Experimental setup for the generation and detection of tunable THz pulses. The zone plate producing a pulse train is explained in the inset. ZP – zone plate; L – lens; ΔT – time delay; OAP – off axis parabolic mirror; WG – wire grid; RTM – roof top mirror; DAST – nonlinear crystal: dimethyl amino 4-N-methylstilbazolium tosylate

is the group velocity of the optical pulse. Therefore the zone plate causes a time delay $N \times \Delta t$ in that portion of the beam which passes through N slides. This leads to a pulse train of repetition rate $1/\Delta t$.

Three zone plates of glass thickness 112, 160 and 220 µm, respectively, were used to cover a large frequency range. This yields a time delay Δt of 186, 266 and 366 fs, respectively. Optical rectification is achieved by using the highly nonlinear organic crystal, dimethyl amino 4-N-methylstilbazolium tosylate (DAST) [20]. The pulse trains are focused on the rear side of the crystal to a diameter of 25 µm. Since phase matching between the FIR and the optical pulses is not possible, the extent of the THz radiation source is limited to the coherence length of several micrometers. Because this dimension is small compared to the FIR wavelength of about 100 µm, the radiation source behaves like an oscillating dipole which emits into a large solid angle. An off-axis parabolic mirror collimates and directs the FIR signal to a Michelson interferometer in a Martin–Puplett configuration [21]. An optical path delay of 19 mm corresponds to a time delay of ~ 60 ps and yields a spectral resolution of 15 GHz. The interferogram is measured with a liquid He cooled, photoconducting Ge:Ga detector sensitive to wavelengths between 50 and 140 µm. Numerical Fourier transformation yields the power spectrum of the THz interferogram. In order to prevent absorption by water vapor in air, the entire FIR setup is immersed in an airtight enclosure and flushed with dry nitrogen or argon gas.

The detected interferogram of a single THz-pulse and its spectrum are shown in Fig. 2. Theoretically, the spectrum should cover the range from 1 to 12 THz. Due to the response characteristics of the Ge:Ga detector the measured signal is narrower and resembles essentially the detectivity curve shown in the inset in Fig. 2b. The oscillatory structure in the wings of the interferogram leads to several dips in the frequency spectrum that vary in depth with the relative humidity of air (see Fig. 2b). We therefore attribute these spectral features to the absorption by residual water vapor inside the enclosure [22].

Figure 3a shows the measured interferogram of a THz pulse generated by a sequence of 5 optical pulses. The inset displays the measured cross-correlation of a single optical pulse and the pulse train with a repetition rate of 3.4 THz. The peak of the corresponding spectrum (Fig. 3b) is at 3.4 THz and its FWHM of approximately 0.6 THz is drastically narrowed in comparison to the THz signal of a single optical pulse. The

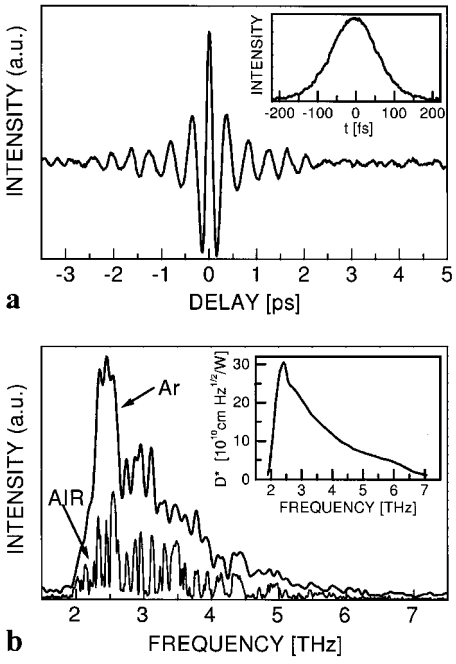

Fig. 2. a THz interferogram of a rectified optical pulse. Its autocorrelation is shown in the inset. **b** Fourier transform of the above signal. For comparison, the Fourier transform of an interferogram detected in air of 40% relative humidity is shown in the lower curve. The inset displays the detectivity curve of the Ge:Ga photoconductor [21]

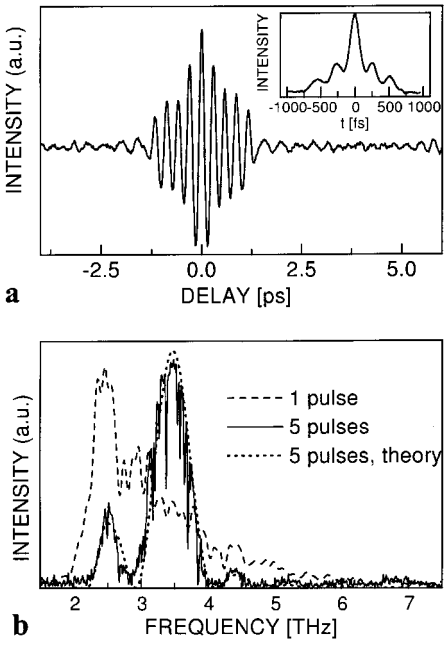

Fig. 3. a THz interferogram of a rectified optical pulse train. Inset: Cross-correlation of the pulse train consisting of 5 pulses. **b** Power spectrum of (**a**) (*solid curve*) compared to the spectrum of a single pulse (*dashed curve*) and theory (*dotted line*)

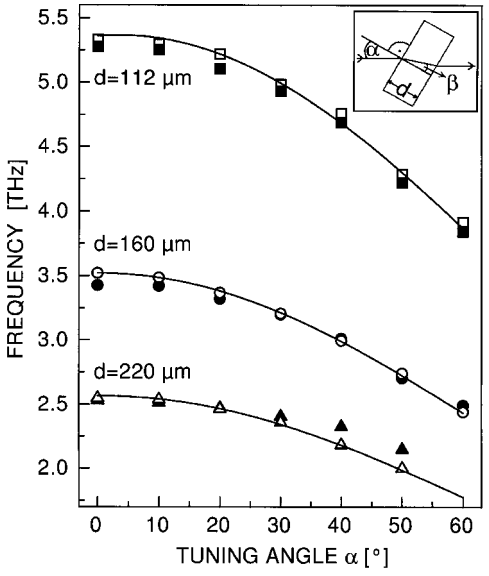

Fig. 4. Experimental data and theoretical tuning curve for the peak frequency of the THz radiation vs. the tilt angle for three different zone plates. The inset shows schematically a single glass plate of thickness *d*. The full symbols represent the raw data and the hollow symbols take into account the spectral characteristics of the Ge:Ga detector

second peak at 2.5 THz is a combined effect of the enhanced detector sensitivity around 2.4 THz and the finite number of laser pulses in the pulse train leading to higher order Fourier components in the spectrum.

As shown in Fig. 3b, the theory is in good agreement with the experimental data. The calculations took into account the non-Gaussian envelope of the optical pulse train (finite number of pulses in the pulse train) and the detectivity curve of the Ge:Ga detector. It should be noted that pulse trains with more pulses will narrow the spectral bandwidth further.

The tunability of the THz radiation is shown in Fig. 4. The FIR radiation is generated by trains of 5 optical pulses. The tuning curves were obtained with three different zone plates, consisting of 4 glass slides of thickness 112, 160 and 220 µm respectively. The full symbols represent the raw data and the hollow symbols take into account the spectral characteristics of the Ge:Ga detector. By tilting the zone plates up to an angle of 60°, we tuned the peak frequency of the THz pulses from 2.0 to 5.3 THz. The calculated tuning curve is in good agreement with the experimental data. The detected average THz power was ~ 100 nW for a single pulse and ~ 20 nW for a pulse train. This corresponds to a THz peak power of about 10 mW and 0.5 mW, respectively.

In summary, we have presented a new and simple method for the generation of tunable narrow bandwidth FIR pulses via the rectification of optical pulse trains. The THz power can be increased by several orders of magnitude if amplified femtosecond laser pulses are used. Furthermore, the spectral range of the generated FIR radiation is easily extended to both lower and higher frequencies by the use of glass slides of different thicknesses and proper duration of the optical pulses. The linewidth can be narrowed further by increasing the number of zones. The shape of the THz pulses as well as their chirp can be arbitrarily designed by variation of the geometrical parameters of the zone plate.

To our knowledge, our work demonstrates the first application of a zone plate for the generation of a pulse train with a THz repetition rate. For the first time, optical rectification of these pulse trains in a nonlinear crystal is demonstrated leading to a substantial narrowing of the spectral bandwidth. The easy handling of the whole system and the large tuning range make it ideal for time resolved spectroscopy.

Acknowledgements. We thank Prof. E. Gornik for supplying the Ge:Ga detector and R. Bischof for experimental help. This work has been supported by the Jubilaeumsfonds der Oesterreichischen Nationalbank, project #5059, and by the Fonds zur Förderung der Wissenschaftlichen Forschung, project #P10065.

References

1. D.H. Auston, K.P. Cheung, P.R. Smith: Appl. Phys. Lett. **45**, 284 (1984)
2. P.R. Smith, D.H. Auston, M.C. Nuss: IEEE J.of Quantum Electron. **QE-24**, 255 (1988)
3. P.C.M. Planken, M.C. Nuss, I. Brener, K.W. Goossen: Phys. Rev. Lett. **69**, 3800 (1992)
4. K. Leo, J. Shah, E.O. Göbel, T.C. Damen, S. Schmitt-Rink, W. Schäfer, K. Köhler: Phys. Rev. Lett. **66**, 201 (1991)
5. C. Waschke, H.G. Roskos, R. Schwedler, K. Leo, H. Kurz, K. Köhler: Phys. Rev. Lett. **70**, 3319 (1993)
6. X.-C. Zhang, B.B. Hu, J.T. Darrow, D.H. Auston: Appl. Phys. Lett. **56**, 1011 (1990)
7. A. Bonvalet, M. Joffre, J.L. Martin, A. Migus: Appl. Phys. Lett. **67**, 2907 (1995)
8. D.H. Auston, K.P. Cheung, J.A. Valdmanis, D.A. Kleinmann: Phys. Rev. Lett. **53**, 1555 (1984)
9. K.H. Yang, P.L. Richards, Y.R. Shen: Appl. Phys. Lett. **19**, 320 (1971)
10. X.-C. Zhang, X.F. Ma, T.-M. Lu, E.P. Boden, P.D. Phelps, K.R. Stewart, C.P. Yakymyshyn: Appl. Phys. Lett. **61**, 3080 (1992)
11. T.J. Carrig, G. Rodriguez, T.S. Clement, A.J. Taylor, K.R. Stewart: Appl. Phys. Lett. **66**, 121 (1995)
12. A.S. Weling, B.B. Hu, N.M. Froberg, D.H. Auston: Appl. Phys. Lett. **64**, 137 (1994)
13. I. Brener, P.C.M. Planken, M.C. Nuss, L. Pfeiffer, D.E. Leaird, A.M. Weiner: Appl. Phys. Lett. **63**, 2213 (1993)
14. P.C.M. Planken, I. Brener, M.C. Nuss, M.S.C. Luo, S.L. Chuang: Phys. Rev. B **48**, 4903 (1993)
15. K. Kawase, M. Sato, T. Taniuchi, H. Ito: Appl. Phys. Lett. **68**, 2483 (1996)
16. M. van Exter, Ch. Fattinger, D. Grischkowsky: Opt. Lett. **14**, 1128 (1989)
17. B.B. Hu, M.C. Nuss: Opt. Lett. **20**, 1716 (1995)
18. J. Zielbauer, M. Wegener: Appl. Phys. Lett. **68**, 1223 (1996)
19. Y.R. Shen: *The Principles of Nonlinear Optics* (Wiley, New York 1984), p 57
20. J.W. Perry, S.R. Marder, K.J. Perry, E.T. Sleva, C. Yakymyshyn, K.R. Stewart, E.P. Boden: *Nonlinear Optical Properties of Organic Materials IV* 2, Proc SPIE 1560, 302 (1991)
21. D.H. Martin, E. Puplett: Infrared Physics **10**, 105 (1969)
22. F. Matsushima, H. Odashima, T. Iwasaki, S. Tsunekawa, K. Takagi: Journal of Molecular Structure **352**/**353**, 371 (1995)
23. P.R. Bratt: in *Semiconductors and Semimetals*, (R.K. Willardson, A.C. Beer, eds.), Vol. 12, Chap. 2, Fig. 34, p. 106 (Academic Press, New York 1977)