

Spectral filtering of femtosecond laser pulses by interference filters

R. Szipöcs^{1,2}, A. Köházi-Kis², P. Apai¹, E. Finger³, A. Euteneuer³, M. Hofmann³

¹Research Institute for Solid State Physics and Optics, P.O. Box 49, H-1525 Budapest, Hungary

²R&D Ultrafast Lasers Kft, Konkoly Thege 29-33, H-1121 Budapest, Hungary

³Fachbereich Physik, Philipps-Universität, Renthof 5, D-35032 Marburg, Germany

Received: 1 October 1999/Revised version: 15 March 2000/Published online: 24 May 2000 – © Springer-Verlag 2000

Abstract. Phase properties of optical thin film interference filters are discussed from the aspect of their usage for phase-error free wavelength separation of broadband femtosecond laser pulses. It is shown that both transmissive or reflective interference filters with high contrast ratios exhibit high cubic phase shifts on transmission or reflection, respectively, causing intolerable distortion in the temporal pulse shape. We show, however, that high efficiency wavelength separation of broadband femtosecond laser pulses can be achieved by using low contrast, properly designed reflective optical interference filters directly built into the cavity of the broad spectrum, femtosecond pulse lasers or parametric oscillators. For demonstrative purposes, we implemented the idea for a Kerr-lens mode-locked Ti:sapphire laser, and obtained two-color, inherently synchronized, unchirped, femtosecond pulse outputs from a single laser oscillator.

PACS: 45.15.Eq; 42.25.Bs; 42.40.Pa; 42.60.Da; 42.65k; 42.79.Bh; 42.90.V

Broadly tunable laser systems form the basis of modern optics. In particular, Ti:sapphire-based laser systems are very convenient and powerful sources of coherent light, tunable over a broad bandwidth. The discovery of ultrafast pulse generation by self-mode-locking [1] in these systems was a breakthrough for many fields of applications. The high peak powers of ultrashort pulses are exploited in a variety of nonlinear optical techniques [2] whereas the short pulse durations are important for time-resolved spectroscopy [3]. Nonlinear optical spectroscopy profited from the possibility of tuning the emission spectrum of the pulsed lasers resonant to any optical transition in the investigated material [4].

In many spectroscopic applications, the availability of two or more pulse trains with independently tunable different spectra is a basic requirement. Typical examples are pump probe experiments on organic materials [5] and nondegenerate four-wave mixing experiments using pulse trains, either with different spectral positions [6] or with different spectral widths [7]. These techniques require a femtosecond laser system offering multiple, highly synchronized outputs with independently tunable spectra.

However, the simultaneous generation of multiple pulses with different wavelengths but controllable temporal pulse separation up to now has still required rather complicated techniques. One possibility is the extracavity manipulation of the pulses by spectral filters [7] or Fourier-transform techniques [8]. The advantage of these methods is that the jitter between the different pulses can be zero, because they all stem from the same original pulse. The main disadvantage of using high contrast spectral filters is the distortion of the temporal shape of a femtosecond laser pulse. In Fig. 1, the computed transmittance and group delay on transmission are plotted for a typical thin film realization of a Fabry-Perot-type, bandpass (transmission) interference filter. In Fig. 2, the computed reflectance and group delay on reflection is plotted for a possible thin film realization of a (reflective) notch filter. In general, we found that each realization of both types of spectral filters show a huge (positive or negative) cubic phase shift around the filters' central frequencies, distorting the temporal shape of a femtosecond laser pulse being transmitted or reflected by these filters. This physical behavior of optical interference filters can be understood on the basis of the theoretical works of Tikhonravov [9] and Verly and Dobrovolsky [10], who showed that "the phase factor will vary rapidly in all small intervals near the amplitude reflectance zeros," i.e., at around the central frequency of an interference filter. They also found that "... the physical phase exhibits a phase shift of π rad at the wavenumbers corresponding to the transmittance maxima. The deeper and narrower the passband, the sharper the shifts." In other words: the narrower and deeper the transmittance band of a filter, the higher the group delay introduced by the filter upon transmission.

For the simultaneous generation of multiple pulses with different wavelengths but controllable temporal pulse separation, several groups synchronized the outputs of two separate cavities by coupling them via a common gain medium [11–13]. In these complicated two-color lasers, the jitter between the two pulse trains can be reduced to less than 2 fs [13]. However, these complicated laser setups are very difficult to align and operate; thus, they are rarely used in ultrafast spectroscopy laboratories.

Considering all the difficulties and disadvantages listed above, we came to the following conclusion: because of

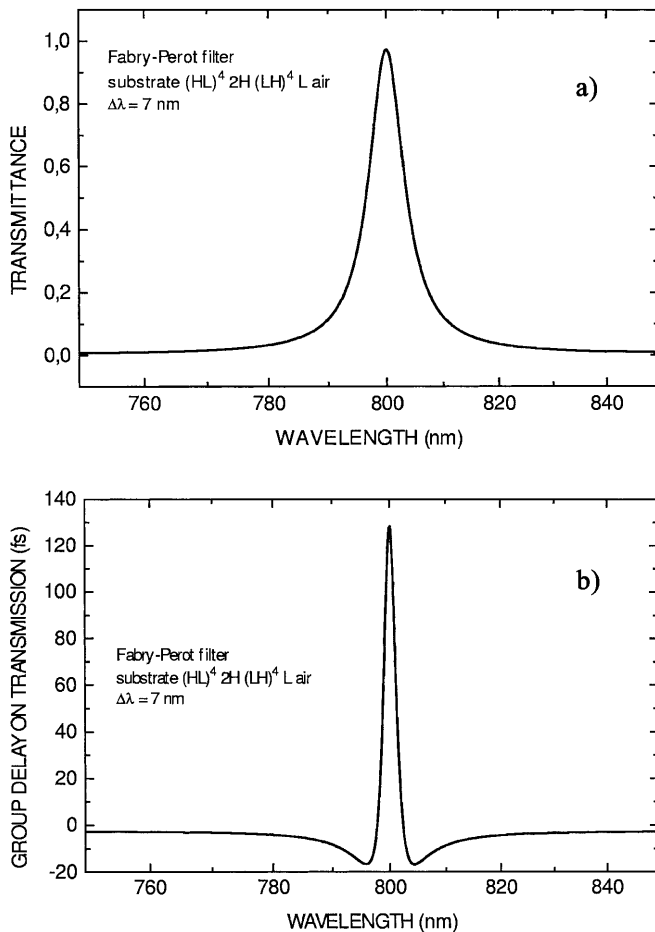


Fig. 1. **a** Computed transmittance and **b** group delay on the transmission of a thin film realization of a Fabry-Perot type interference filter having the structure of substrate (HL)⁴ 2H (LH)⁴ L air, where H and L denote quarterwave thick layers of the high ($n_H = 2.31$) and low ($n_L = 1.45$) refractive index materials at the central wavelength of 800 nm

the basic theoretical reasons discussed above, it is impossible to design optical interference filters for the wavelength separation of ultrabroadband spectrum femtosecond pulses with high contrast ratios and low phase distortion at the same time. In femtosecond pulse lasers, however, only a few percent of the intracavity field is coupled out by some partially transmitting laser mirrors balancing the gain and the loss in the laser cavity. During our studies we found, however, that it is possible to design optical thin film interference filters with low contrast ratios and with moderate phase distortions. Placing such low contrast filters into ultrabroadband femtosecond pulse laser resonators or optical parametric oscillators, we can build multicolor femtosecond laser sources with inherently synchronized outputs and no energy loss because of extracavity spectral filtering. We implemented the idea in a Kerr-lens mode-locked Ti:sapphire laser: we generated ultrashort and thus broadband intracavity pulses and used two separate, narrow band output couples to extract two independent parts of the pulse spectrum. The pulses are inherently synchronized since the two resulting pulse trains originate from the same internal pulses.

The generation of ultrashort pulses requires the precise control of intracavity dispersion. Initially, a fused silica prism

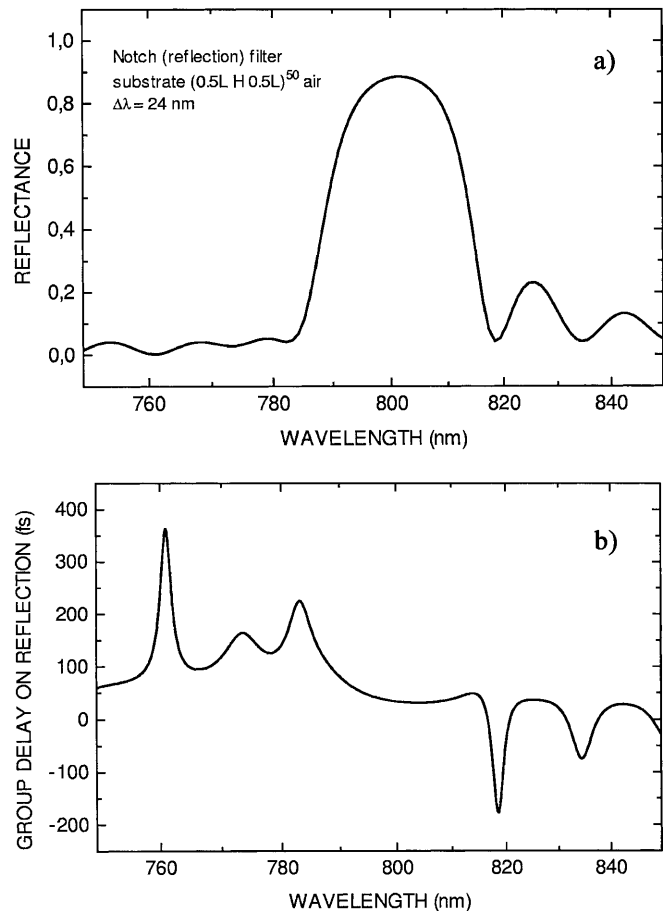


Fig. 2. **a** Computed reflectance and **b** group delay on reflection of a thin film realization of a typical realization of a (reflective) notch interference filter having the structure of substrate (0.5L H 0.5L)⁵⁰ air, where H and L denote quarterwave thick layers of the high ($n_H = 1.5$) and low ($n_L = 1.45$) refractive index materials at the central wavelength of 800 nm

pair was used. Later, chirped mirrors [14] improved the dispersion compensation, yielding 11-fs pulses without the use of prisms [15]. The tunability also profited from the development of chirped mirrors. Special ultrabroadband mirrors have been developed for prism-controlled oscillators. These mirrors have a broader reflectance band and improved dispersion properties in comparison to usual dielectric highreflectors [16]. This concept of hybrid dispersion control, combining a pair of quartz prisms with broadband chirped mirrors, is the basis of our experiment. Figure 3 shows the setup of the linear resonator. The 4-mm-long (path length) Ti:sapphire crystal ($\alpha = 6.0 \text{ cm}^{-1}$) is pumped with 3.5 W at 532 nm from an intracavity frequency-doubled Nd:YVO₄ laser. The intracavity dispersion is controlled by two chirped mirrors (CM1, CM2) and a pair of quartz prisms (P1, P2). The two output couplers (OC1, OC2) are installed exactly at Brewster angle to avoid disturbing etalon effects. Their reflectivity and dispersion spectra are shown in Fig. 4. The output spectra as well as the intracavity pulse spectrum are measured with a fiber-coupled spectrometer. The pulse durations, after extracavity pulse compression with additional chirped mirrors, are determined by fringe-resolved autocorrelation measurements.

First, we optimized the cavity without the Brewster output couplers: at 3.5-W pumping, the laser delivers < 12 fs

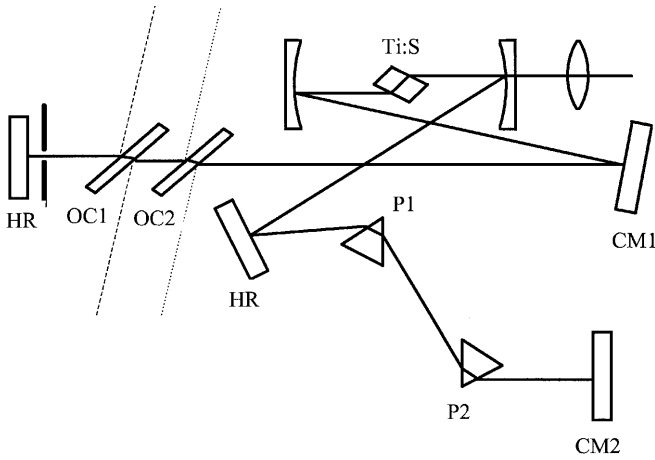
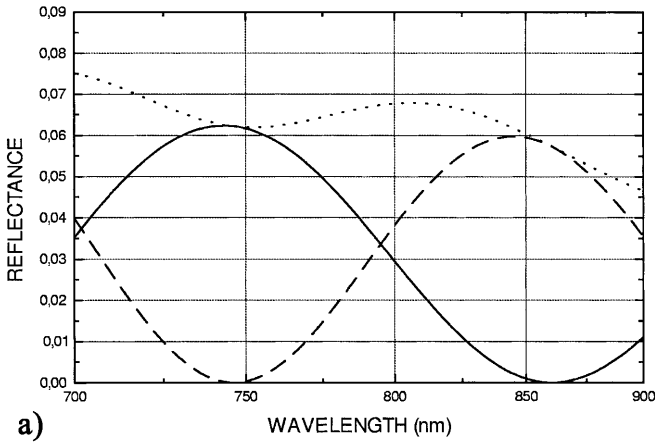
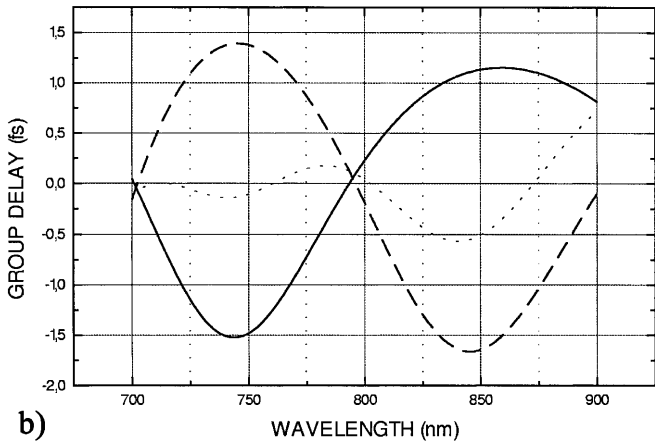


Fig. 3. Schematic sketch of the cavity setup



a)



b)

Fig. 4. a Reflectivity and **b** group delay on the transmission of shortwave output coupler (continuous lines) and longwave output coupler (dashed lines). The overall values are also plotted (dotted lines)

pulses with 550 mW mode-locked output power at 790 nm when using a $T = 16\%$ output coupler. Then, this output coupler is replaced by a high reflector (HR), and the two Brewster OCs are inserted. The positive dispersion of their two quartz substrates is compensated by moving one of the prisms. The vertical slit is translated horizontally to account for the small shift of the beam. There is no need for further alignments.

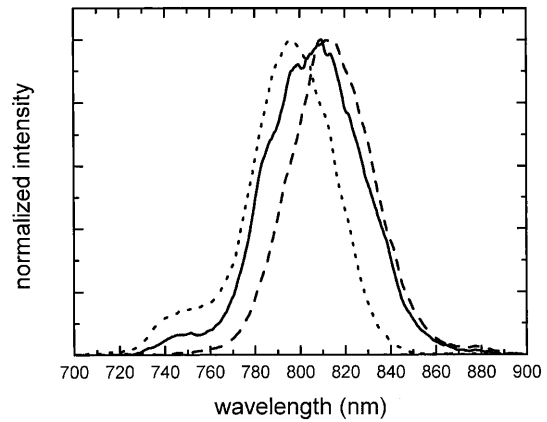


Fig. 5. Two-color operation: the two extracavity spectra coupled out by the long wave OC (dashed line) and the short wave OC (dotted line) together with the intracavity spectrum (solid line) [17]

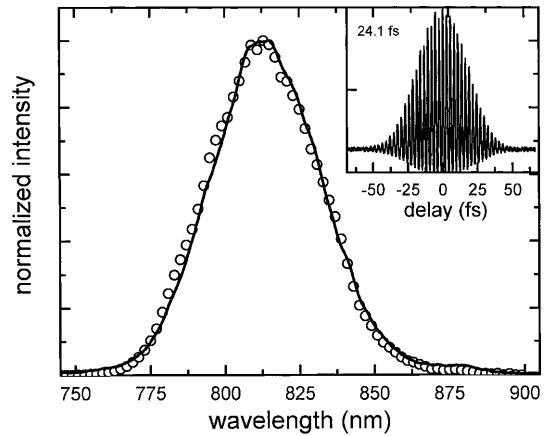


Fig. 6. Pulse spectrum from the long wave OC: measured (solid line) and calculated by multiplying the intracavity spectrum by the reflectivity of the long wave OC (circles). Inset: respective fringe resolved autocorrelation trace

An intracavity pulse spectrum, taken from light scattered off the high reflector (HR) in the longer arm of the cavity, is shown in Fig. 5 as a solid line. Its full-width-at-half-maximum (FWHM) is 23.4 THz^1 . The output powers are 100 mW (OC1) and 40 mW (OC2) plus the respective values at the back sides of the OCs. The spectra of the two outcoupled pulse trains are shown in Fig. 5 as a dashed and a dotted line. They are centered around the intracavity spectrum and their central wavelengths are separated by 17 nm. The spectral shape of the pulses is best fitted with a Gaussian shape rather than a sech^2 shape. However, the resulting pulse shapes in the actual setup are determined by the reflection properties of the OCs. In Fig. 6 the measured pulse spectrum from the long wave output coupler (solid line) is compared with a spectrum (circles) calculated by multiplying the intracavity spectrum (Fig. 5) with the reflectivity of the corresponding output coupler (Fig. 4a). The perfect correspondence shows that for a given intracavity spectrum the output spectra can be

¹ The intracavity pulse spectrum is measured from scattered light resulting in a poor signal-to-noise ratio. The spectrum is best fitted with a Gaussian pulse shape of 23.4 THz FWHM, corresponding to a minimum pulse duration of 18.8 fs.

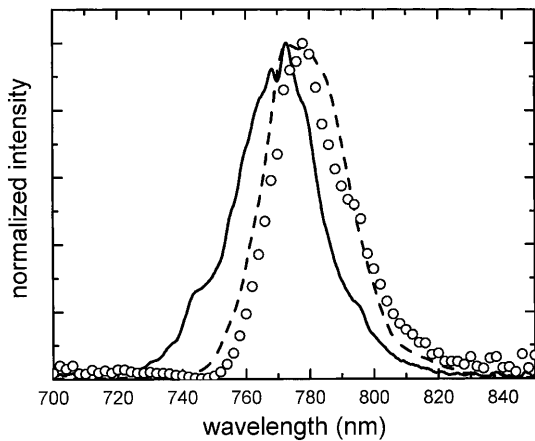


Fig. 7. Single-color operation without short wave OC: the measured extracavity spectrum (*dashed line*) is well reproduced (*circles*) by multiplying the intracavity spectrum (*solid line*) by the reflectivity of long wave OC

tailored by designing the respective reflection profiles of the OCs. The fringe-resolved autocorrelation measurements yield pulse durations in the two pulse trains of 24.1 fs (inset Fig. 6) and 24.5 fs, respectively. With the spectral bandwidths of 19.7 THz and 20.3 THz, we get time-bandwidth products of 0.475 and 0.497, respectively. The deviation from the limit of 0.441 for Gaussian pulses is due to the slightly non-Gaussian spectral shapes and some residual chirp. The latter is caused by the fact that the dispersion of the Ti:sapphire crystal is compensated mainly in the resonator arm with the prism pairs, whereas the OCs are placed in the other arm. Thus, the extracavity pulses are chirped since they are coupled out at a position of the cavity where the intracavity pulses are chirped. The same experiment was performed with less negative dispersion in the cavity, i.e., more prism material in the beam path. In this case, the better balance of the intracavity dispersion leads to time-bandwidth products very close to 0.441. However, the resulting pulses outside and inside the cavity are spectrally narrower and thus longer (e.g., 12.8 THz at the long wave OC corresponding to 34.5 fs).

The spectral distribution of losses in the cavity is an important point for the operation of our setup. Figure 7 shows the intracavity spectrum (solid) and the extracavity spectrum at the long wave OC (dashed) when the short wave OC is removed. The extracavity spectrum is then not at the desired wavelength although it is still shifted with respect to the intracavity spectrum. The intracavity spectrum as well as the spectrum coupled out by the long wavelength OC become narrower and shift to shorter wavelengths when the short wavelength OC is removed. The losses, introduced at longer wavelengths by the residual OC, force the laser to run at shorter wavelengths. This indicates that for multicolor operation the overall losses, introduced by all the OCs, must be distributed symmetrically over the desired spectral range. Tuning one of the OCs would thus require symmetrical tuning of some additional loss element. Further studies have to show whether this problem of spectral loss balancing can be overcome, e.g., by the outcoupling of lower powers. Furthermore, in our present, simple, linear cavity setup each OC leads to two outputs (see Fig. 3). The second output is usually not

needed and thus acts as an additional wavelength-dependent loss. This additional loss might be circumvented in a ring-cavity configuration.

In summary, we have suggested that low contrast optical interference filters, e.g., Brewster angled output couplers can be well suited for building new and simple lasers or parametric oscillators for multicolor operation. We implemented the idea in an ultrafast, single cavity, Kerr-lens mode-locked Ti:sapphire laser with two output couplers. First experiments with two output couplers demonstrate the potential of such a new laser: two synchronous pulse trains with spectra separated by 17 nm are generated. The pulses are nearly transform limited with pulse durations of about 24 fs. The intracavity pulse spectrum is broader than the output spectra and spectrally centered between them. Our results demonstrate that our new simple concept exhibits the desired properties: Two spectrally different but temporally synchronized pulse trains are produced. The individual tunability of the two outputs could be realized with laterally structured dielectric coatings and translation of the respective OC. In principle any desired pulse shapes could also be generated using OCs with corresponding design of the reflection profile. The main results of this paper were first presented at the Ultrafast Optics 99 conference [17].

Acknowledgements. The authors gratefully acknowledge helpful discussions with W.W. Rühle. The work was supported by the German-Hungarian project WTZ UNG-051-96 (D-85/96) and by the OTKA Science Foundation of Hungary under grants T 029578 and T 022563.

References

1. D.E. Spence, P.N. Kean, W. Sibbett: *Opt. Lett.* **16**, 42 (1991)
2. Y.R. Shen: *The Principles of Nonlinear Optics* (Wiley, New York 1984)
3. W. Kaiser (ed.): *Ultrashort Laser Pulses – Generation and Application* (Springer, Berlin Heidelberg 1988, 1993)
4. S. Mukamel: *Principles of Nonlinear Optical Spectroscopy* (Oxford University Press, New York 1995)
5. S.H. Ashworth, I. Hasche, M. Woerner, E. Riedle, T. Elsaesser: *J. Chem. Phys.* **104**, 5761 (1996)
6. D.S. Kim, J.Y. Sohn, J.S. Yang, Y.K. Ahn, K.J. Yee, Y.D. Jho, S.C. Hohng, D.H. Kim, W.S. Kim, J.C. Woo, T. Meier, S.W. Koch, D.H. Woo, E.K. Kim, S.H. Kim, C.S. Kim: *Phys. Rev. Lett.* **80**, 4803 (1998)
7. S.T. Cundiff, M. Koch, W.H. Knox, J. Shah, W. Stolz: *Phys. Rev. Lett.* **77**, 1107 (1996)
8. A.M. Weiner, J.P. Heritage, E.M. Kirschner: *J. Opt. Soc. Am. B* **5**, 1563 (1988)
9. A.V. Tikhonravov: *Appl. Opt.* **32**, 5417 (1993); A.V. Tikhonravov, P.W. Baumeister, K.V. Popov: *Appl. Opt.* **36**, 4382 (1997)
10. P.G. Verly, J.A. Dobrowolski: *Appl. Opt.* **29**, 3672 (1990)
11. J.M. Evans, D.E. Spence, D. Burns, W. Sibbett: *Opt. Lett.* **18**, 1074 (1993)
12. D.R. Dykaar, S.B. Darack: *Opt. Lett.* **18**, 634 (1993)
13. A. Leitenstorfer, C. Fürst, A. Laubereau: *Opt. Lett.* **20**, 916 (1995)
14. R. Szipöcs, K. Ferencz, Ch. Spielmann, F. Krausz: *Opt. Lett.* **19**, 201 (1994); R. Szipöcs, A. Köhási-Kis: *Appl. Phys. B* **65**, 115 (1997)
15. R. Szipöcs, F. Krausz: U.S. Patent No. 5 734 503; A. Stingl, C. Spielmann, F. Krausz, R. Szipöcs: *Opt. Lett.* **19**, 204 (1994)
16. E.J. Mayer, J. Möbius, A. Euteneuer, W.W. Rühle, R. Szipöcs: *Opt. Lett.* **19**, 528 (1997)
17. R. Szipöcs, A. Köhási-Kis, P. Apai, E. Finger, A. Euteneuer, M. Hofmann: Technical Digest of the Ultrafast Optics 99 Conference, Monte Verita, Ascona, Switzerland, 11–16 July 1999, pp. 74–77 (1999)