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Feedback-controlled femtosecond pulse shaping

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Abstract. We describe the experimental implementation of feedback-optimized femtosecond laser pulse shaping. A frequency-domain phase shaper is combined with different pulse characterization methods and appropriate optimization algorithms to compensate for any phase deviation. In particular, bandwidth-limited, amplified laser pulses are achieved by maximizing the second-harmonic generation (SHG) of the shaped laser pulses with the aid of an evolutionary algorithm. Real-time measurement of the absolute phases is achieved with spectral interferometry where the reference pulse is characterized by FROG, the so-called TADPOLE method. Using the complete electric field as feedback, arbitrary laser pulse shapes can be optimally generated in two different ways. First, a local convergence algorithm can be used to apply reliable and accurate spectral chirps. Second, an evolutionary algorithm can be employed to reach specific temporal profiles.

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Femtosecond pulse shaping has become an increasingly important technology in the last years with numerous applications in many different fields. In general, fundamental laser-induced processes in physics, chemistry or biology can not only be studied but even actively controlled and optimized if ultrashort laser pulses with specific intensity and phase profiles are available.

After the first work on ps pulse shaping by Heritage et al. [1], tailoring fs laser pulses was introduced by Weiner et al. [2, 3]. Programmable pulse shapers have been developed which make use of a liquid crystal display (LCD) spatial light modulator (SLM) [2–4] within a zero dispersion compressor. Non-pixelated liquid crystal devices [5], acousto-optic modulators (AOM) [6, 7] as well as deformable mirrors [8] have also been reported as filtering devices. In principle, almost arbitrary pulse shapes can be produced in such a spectral phase/amplitude modulation setup. However, additional phase terms from optical elements within the pulse shaper may reduce the fidelity of the output pulse shape, as will be discussed in detail later. It would be extremely helpful if a method of real-time characterization of the complex pulse shapes and automated correction for any errors existed.

Recently, the powerful method of “self-learning” or “adaptive” pulse shaping has been introduced. A suitable computer algorithm uses direct feedback from the experiment in order to find the “best” AOM or LCD pattern in a given optimization problem [9, 10]. Automated fs laser pulse compression has been achieved for unamplified laser pulses by using the efficiency of second-harmonic generation (SHG) as feedback [11, 12]. In the case of amplified laser pulses, SHG [13] as well as spectral blueshifting [14] have been used for automated generation of bandwidth-limited laser pulses without requiring any knowledge of the input pulse shape. If arbitrary, specifically designed laser pulses are desired, the feedback signal has to be generalized accordingly. As a first realization of this scheme, intensity cross-correlation of unamplified laser pulses was used to generate the feedback signal [15]. This offers the advantage that no measurement of the input pulse has to be made, but with the disadvantage that the intensity correlation does not yield the complete phase information. In another approach, spectral interferometry [16, 17] of unamplified laser pulses has been used to characterize and improve the relative phase introduced by the pulse shaper setup [18].

In practice, however, laser pulses with an accurately shaped absolute phase and intensity profile are desired. We therefore combine the TADPOLE method (temporal analysis by dispersing a pair of light e-fields) [19], which is suitable for complete characterization of the electric field, and a frequency-domain pulse shaper to generate feedback-optimized almost arbitrary fs laser pulses. In this paper, we cover the following three aspects of automated fs pulse shaping: generation of bandwidth-limited laser pulses by SHG maximization with an evolutionary algorithm, precise realization of specific spectral phase modulations (spectral chirp) with a local convergence algorithm and tailoring of specific temporal intensity profiles with an evolutionary algorithm.

1 The experimental techniques

In these experiments on automated pulse shaping we combine the following devices and techniques as can be seen in Fig. 1: a fs laser system, a frequency-domain pulse shaper, different laser pulse characterization methods and suitable computer algorithms for the pulse shaper control which make use of appropriate feedback from the pulse characterization.

Our laser system consists of a home-built Ti:sapphire oscillator and a commercial chirped-pulse amplification (CPA) system, capable of producing 80 fs, 1 mJ pulses at 800 nm.

The pulse shaper is based on a design of Weiner et al. [3]. We have described our setup previously [13]. The fs pulse spectrum is dispersed and recollimated within a zero-dispersion compressor, consisting of a pair of 1800 lines/mm gratings and a pair of 80-mm-focal-length plano-cylindrical lenses. In the Fourier plane of the system, the spatially separated spectral components can be manipulated by a phase and/or amplitude mask. We use a one-layer liquid-crystal display (LCD), the spatial light modulator SLM-128 by Cambridge Research & Instrumentation, suitable for phase modulation in 128 independent pixels across the pulse spectrum.

As has been remarked in the introduction, however, unwanted additional phase terms may originate within the pulse shaper. If accurate pulse shaping is required, one must therefore usually consider the following issues. First of all, there are fundamental limitations to this method of pulse shaping. The pixelation of the LCD filter masks and the limited phase range accessible by each pixel define a "time window" outside of which pre- and post-pulses may emerge. The presence of unmodulated contributions from light missing the active area of the modulator leads to additional contributions around time zero. Effects from diffraction off the mask (which are fundamental to all Fourier pulse-shaping techniques) may introduce spatial chirp [4, 20, 21]. There are also optical deficiencies such as lens aberrations, material dispersion leading to temporal pulse broadening and limited spectral transmission due to non-ideal transmission/reflection efficiency curves of the optical elements. Alignment issues are also very critical. One must adjust the distances of the individual elements with respect to each other and with respect to the optical axis, as well as adjust their tilt. Another difficulty is added because all optical elements in the pulse shaper have some cylindrical symmetry which has to be taken care of; i.e., the lines of the grating, the axes of the lenses, the axes of the half-wave plates as well as the axis of the LCD all have to be aligned accordingly. Further on, there are two calibrations which have to be accurate, namely the assignment of the LCD voltages to the corresponding phase responses, and the registration of the pixel number with the corresponding wavelength range. One must also keep in mind that the input laser pulses may not be bandwidth-limited but could exhibit different temporal/spectral and spatial pulse shapes.

Considering all these difficulties it is essential to use adequate pulse characterization methods. We mainly use interferometric autocorrelation, SHG-FROG and TADPOLE, depending on the context of the measurements. If nearly bandwidth-limited laser pulses are to be analyzed, standard interferometric autocorrelation in a nonlinear crystal offers great sensitivity. Slight deviations from the ideal unchirped case are readily visible in an increased FWHM or in additional structure in the wings of the autocorrelation trace.

If automated tailoring of specifically and complex-shaped laser pulses is desired, complete and fast analysis of the pulse shaper output is necessary. We then revert to TADPOLE [19], a non-iterative method for complete characterization of fs laser pulses. The combination of TADPOLE with our pulse shaper is depicted in Fig. 1. A Mach–Zehnder interferometer contains the pulse shaper inside one of the two arms. The other arm provides an unmodulated reference pulse which is characterized by SHG-FROG [22]. The two pulses are then collinearly recombined with an adjustable temporal delay, and the resulting interference spectra are recorded by a multichannel spectrometer which should be carefully calibrated [23]. Analysis of the spectral fringes by a Fourier-transform technique [17] yields the relative phase introduced by the pulse shaper. Together with the absolute phase of the reference pulse one obtains a measurement of the absolute amplitude and phase of the modulated laser pulse. As the analysis of the fringe spectra involves only non-iterative methods, the measurement can be carried out in real-time, i.e., in less than a second. (Since different pulse-shaper settings do not affect the reference, it is sufficient to "update" the reference phase every few minutes.) An advantage of TADPOLE as compared to FROG alone is the speed of the pulse shape recovery. Although current development is aiming at "online" FROG measurements [24], it is in general much faster and easier to implement a direct algorithm as in Fourier spectral interferometry instead of an iterative algorithm as in FROG. Self-referencing spectral interferometry (SPIDER) [25] is another very useful concept of complete fs pulse characterization which could be helpful in combination with pulse shaping. However, this nonlinear technique requires adequate chirping

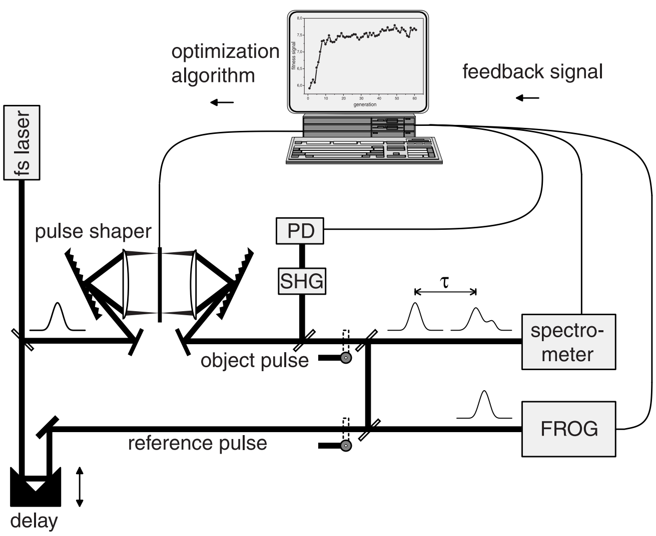

Fig. 1. Experimental setup. A Mach–Zehnder interferometer contains a frequency-domain fs pulse shaper in one of its arms, whereas the second arm provides unmodulated reference pulses with an adjustable temporal delay. The collinear recombination yields spectral fringes which are analyzed by a multichannel spectrometer. Computer-controlled shutters facilitate separate measurements of the object and the reference pulse spectrum. Together with the reference pulse shape determined by FROG, complete real-time characterization of the modulated laser pulses is achieved. Second-harmonic generation (SHG) at the output of the pulse shaper is used as a measure for the pulse duration. Suitable computer algorithms use the different information sources in feedback loops to adaptively phase-shape the laser pulses according to a given target. Imperfections in the pulse shaper setup are thus automatically compensated for

of the (reference) laser pulse, and the fixed amount of chirp may not always be sufficient to cover the whole range of pulse durations produced by the pulse shaper.

The pulse shaper itself is controlled by a suitable computer algorithm (described below) which makes use of some experimental feedback. In the case of automated pulse compression, the SHG efficiency is a suitable choice. The shorter the laser pulses are, the higher is the SHG signal. By maximizing SHG, the laser pulses are therefore compressed. If tailoring of arbitrary pulse shapes is desired, the complete electric field has to be determined instead. We therefore use the result from the TADPOLE measurement as feedback in the optimization algorithm.

The chosen feedback signal is processed either by a local convergence algorithm or by a global evolutionary algorithm, depending on how the feedback behaves with respect to the pulse shaper control. Specifically, the feedback may be called "local" if the change of any LCD pixel voltage introduces a change only in a part of the feedback signal that can be assigned unambiguously to the corresponding LCD pixel. In this case of local feedback (such as the optimization of specific spectral chirps with TADPOLE), the action of the LCD pixels may be optimized individually, and a local convergence algorithm may be used. If, on the other hand, the change of a single LCD pixel voltage changes the feedback signal on the whole (such as in pulse compression by SHG or in the realization of specific temporal profiles), only a global optimization algorithm such as an evolutionary algorithm can be applied.

The local convergence algorithm compares the feedback signal with the desired target and compensates for differences locally. In the case of optimizing a specific spectral chirp, the voltages applied to the LCD are first calculated according to the (approximate) calibrations of the pulse shaper. The actual experimental phase is determined by TADPOLE as described above. It is then compared with the desired target phase, and the LCD voltages are adjusted locally such that the phase difference is compensated for each LCD pixel. If necessary, more than one iteration can be performed.

Evolutionary algorithms are global optimization methods imitating processes of the biological evolution. Two main subgroups of evolutionary algorithms are discussed in the literature: genetic algorithms [26] and evolution strategies [27]. The general working schemes of both are similar, main differences lie in the parameter representation and in the reproduction procedures. Whereas genetic algorithms use binary encoding for the complete genetic information of one individual and reproduction by crossover, evolution strategies use floating-point representations for each parameter to be optimized and the reproduction relies on mutation. A different approach to global optimization is simulated annealing [28], which has also been applied in the context of fs pulse shaping [11, 29, 30]. Our own implementation has been described in detail elsewhere [12] and uses concepts from both types of evolutionary algorithms. Briefly, the 128 voltage values applied to the LCD represent the "genetic configuration" of one "individual", i.e., a single display pattern. The modulated laser pulses are then analyzed by the appropriate feedback signal (SHG or TADPOLE). According to the optimization target, a certain fitness function is calculated (see the following section). The more the output electric field approaches the target pulse shape, the higher is the fitness. After testing all individuals of one generation (randomly initialized), the best ones are selected for reproduction by crossover and mutation procedures. The offspring is usually better adapted to the "environment", leading to an improved laser pulse shape. Repeating this process for many "generations", the average fitness increases until finally the optimization goal of some specific pulse shape is reached.

2 Experiment and discussion

We now combine the experimental techniques described in the preceding section to illustrate the two main issues of automated fs pulse shaping: pulse compression and tailoring of specific pulse shapes.

Even with nowadays rapidly improving laser technology, "truly" bandwidth-limited amplified fs laser pulses are often not readily available. Despite careful alignment of the chirped-pulse amplification (CPA) laser systems there often remains non-negligible higher order dispersion which results in a complex chirp of the laser pulses. To optimize the amplified laser pulses, i.e., to compensate for dispersive terms, we use the evolutionary computer algorithm to maximize the second-harmonic feedback signal. Thereby the laser pulse duration is minimized. Interferometric autocorrelations of amplified laser pulses before and after automated compression are shown in Fig. 2. The wing structure present in the unmodulated laser pulse (Fig. 2a) is clearly removed by the pulse shaper, providing bandwidth-limited pulses of 80 fs duration (Fig. 2b), bounded by the spectrum of our CPA laser system. A detailed FROG analysis of a similar experiment is given in [13]. With an increased bandwidth, the LCD-SLM is capable of producing (unamplified) sub-20-fs laser pulses [11]. Unlike other groups (for example in [21, 31]), we place the pulse shaper subsequent to the amplification stage, thus operating directly with amplified laser pulses. Since we use a phase-only modulator, no additional polarizing optics is required, and the LCD damage threshold is greatly increased as we determined in separate measurements. As compared to our previous publication on pulse compression [13], the maximum input pulse energy could be increased to 800 μJ without risk of damage to the optical components. With an optimized energy throughput of 65%, shaped laser pulses of 520 μJ are available. Our setup offers

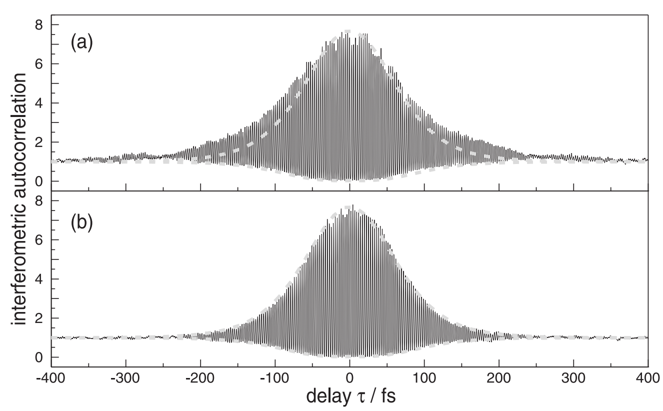

Fig. 2a,a. Interferometric autocorrelations before (**a**) and after (**b**) optimization. The optimal autocorrelation envelope of part **b** has been added to part **a** for comparison several advantages. The pulse shaper acts as a stand-alone device which can be operated with any kind of input laser pulses, no matter whether they originate directly from the laser system, from a frequency conversion unit (for example OPA) or from any other nonlinear process (for example hollow fiber spectral broadening [32]). It is furthermore possible to shape both unamplified and amplified Ti:sapphire fs laser pulses.

In many experiments on atomic or molecular physics as well as photochemistry, however, bandwidth-limited laser pulses offer not the only useful shape. Investigations with chirped laser pulses promise new insight, as well as the prospect to control coherently fundamental laser-induced processes [33, 34]. An asymmetry of the measured experimental signal with respect to zero chirp usually indicates such a chirp dependence rather than a pulse duration (and therefore also pulse intensity) effect of the investigated process. Hence, if measurements are made with varying spectral chirp, one has to ensure that an observed chirp effect, i.e., an experimental response depending on the sign as well as on the quantity of the introduced chirp, is not simply an artifact due to the imperfections of the pulse-shaping device. Consequently, a method has to be introduced that allows for reliable and accurate chirping. We use feedback from the TADPOLE measurement in the local convergence algorithm to correct for deviations.

As an example for the spectral phase optimization we have chosen a linear spectral target chirp of 6000 fs^2 with the given Ti:sapphire oscillator laser spectrum of Fig. 3a. The results of the phase measurements are shown in Fig. 3b. Using the approximate calibration method [3] for the pulse shaper without further optimization, the expected parabolic shape (solid line) is approximately reached (open squares). However, a slight deviation is clearly visible. After the optimization, the measured phase (solid circles) matches the desired phase profile perfectly. Note that with the local convergence algorithm, one or two iterations are usually sufficient to achieve optimal and correct chirping.

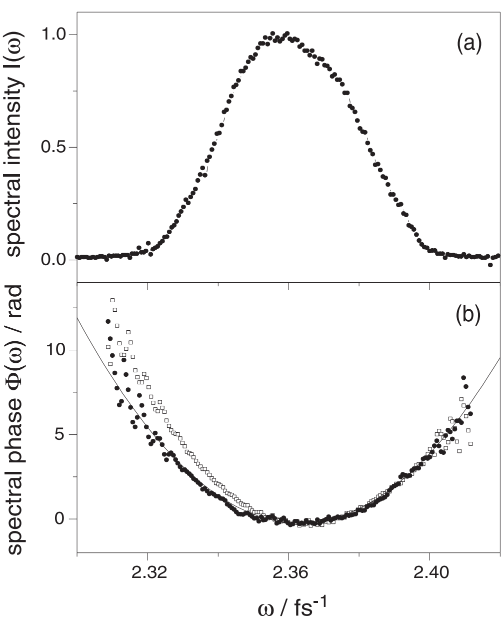

Fig. 3a,b. Laser pulse spectrum (**a**) and phase (**b**) with a linear target chirp of 6000 fs^2 (*solid line*), the measured phase before (*open squares*) and after (*solid circles*) optimization

In order to illustrate that indeed absolute phases can be optimized, we extended the optimization to a whole series of different values of linear chirp, i.e., quadratic phase. In this optimized "chirp scan", we recorded the SHG signal for each of the individual chirp values (Fig. 4). Without optimization (open squares), the maximum of the SHG signal clearly deviates from zero chirp, whereas with optimization (solid circles) the curve is nicely symmetric with respect to zero as is expected for accurate pulse shaping in the case of this SHG "test" experiment. Vice versa, if this optimization method is used and some experimental signal shows an asymmetry with respect to zero chirp, one can be sure that it is indeed a chirp effect. Applying the feedback method offers mainly two advantages. First, one does not have to bother to do very careful calibrations. Although with exact calibrations it may be possible to get high-fidelity pulse-shaping results in many cases, the second and more important advantage of the feedback method is that it also keeps track of time-varying changes in the phase terms and therefore acts as "online" calibration. It opens the possibility to find optimized LCD patterns in cases where a fixed calibration is not sufficient for all possible pulse shapes, i.e., in all cases where the limiting effects as described above depend on the LCD voltages, as well as in the case of phase fluctuations throughout the course of the day.

Exploiting the advantages of TADPOLE, namely the complete real-time characterization of fs laser pulses, online adaptive tailoring of specific temporal pulse shapes can be realized as well. As the spectral bandwidth is limited and fixed, the spectral phase (and therefore the LCD voltages) required to achieve a given temporal pulse shape cannot simply be calculated by a Fourier transform. In general, only approximate realization of the desired pulse shape is possible. Some optimization algorithm has to be used for this purpose. In principle, the spectral phase can be approximately determined a priori and be applied to the pulse shaper after the calculation is done [29, 30]. However, since it is difficult to take into account all the above-mentioned imperfections of the pulse shaper, the fidelity of the output pulse shape may be degraded. We therefore use the experimental TADPOLE measurements as feedback in the evolutionary algorithm.

As a special case of temporal pulse tailoring, we attempted to design laser pulse trains with equally spaced in-

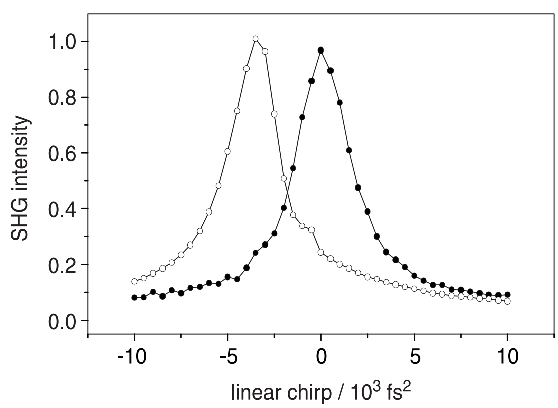

Fig. 4. Chirp scan of second-harmonic generation (SHG) without (*open squares*) and with (*solid circles*) optimization

dividual pulses and given peak heights. Much prior work has been done in the area of pulse train generation [29, 35, 36] or in the related area of spatial Fourier optics (for example see [37, 38]), but without using experimental feedback. A simple method for producing pulse trains by phase-only shaping is based on the so-called maximal length sequences (M-sequences) [35, 39]. There the mask pattern consists of repetitions of a binary phase sequence across the laser spectrum. Each (identical) sequence is divided into p pixels with each pixel being assigned a phase value of either 0 or $\Delta\Phi$ and addressing a frequency interval $\Delta\nu$. The result is a pulse train under a smooth envelope, consisting of p pulses with temporal separations of $1/p\Delta\nu$. As a generalization to that, we wanted to realize a specific temporal envelope, i.e., specific individual peak heights. We therefore used the concept of sequencing the mask pattern with an appropriate number of pixels p but did not use binary phase sequences (or M-sequences). Instead, we let the phase of each pixel of the sequence be determined by the evolutionary algorithm.

Specifically, the measured temporal intensity profile $I(t)$, normalized to unity, is compared with the target shape $I_{\text{target}}(t)$, normalized to unity as well, and a cost functional, the negative of which is the fitness value, is calculated according to

$$\int \left(\frac{I_{\text{target}}(t) - I(t)}{I_{\text{target}}(t) + r} \right)^2 \, \mathrm{d}t$$

with a suitable parameter r. This general fitness function allows continuous tuning between minimization of the relative deviation ($r = 0$) and minimization of the absolute deviation ($r \gg \max\left(I_{\text{target}}(t)\right) = 1$). With $r > 0$ it also avoids division by zero. Qualitatively, minimization of the absolute deviation is sensitive to errors in all parts of the desired pulse shape, whereas minimization of the relative deviation is mainly sensitive to those parts of the target where the absolute signal is desired small (or even zero). Note that the TADPOLE method yields also the temporal phase information. It is therefore possible, in principle, to reach specific adaptively optimized target intensity *and* phase profiles with this method. Although with our one-layer LCD only phases and not amplitudes can be modulated so that there are in general not enough degrees of freedom to provide independent intensity and phase structure, this is not a fundamental limitation to the described feedback-method.

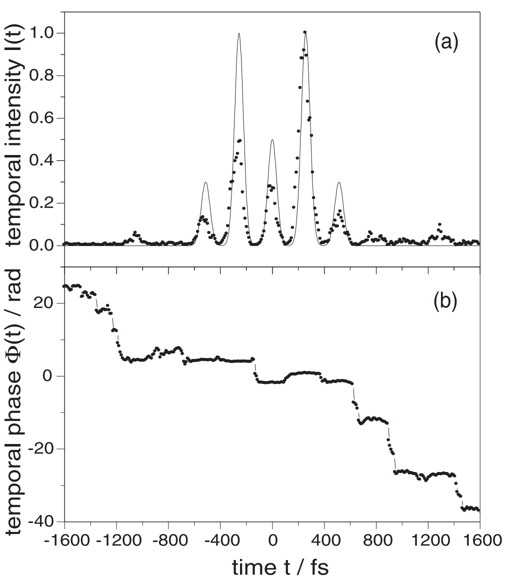

Fig. 5a,b. Temporal intensity (**a**) with the target pulse train (*solid line*) and the measured profile (*solid circles*) as well as the corresponding phase (**b**)

For the demonstration of this temporal pulse tailoring, we chose the target pulse train of Fig. 5a (solid line), consisting of five 80-fs pulses placed at 256 fs separation from each other, but with different peak heights. With the spectral dispersion of our setup, this corresponds to $p = 16$ independent phase values which have to be determined. The evolution of the fitness function ($r = 0.5$), shown in the computer inset of Fig. 1, is qualitatively similar to the fitness curves obtained in the pulse compression experiments [12, 13]. Analysis of the resulting spectral interferogram yields the final temporal pulse shape (Fig. 5a, solid circles). A comparison with the target shape reveals qualitative agreement, but there are also discrepancies which may be explained by the following reasons. Since no amplitude modulation is possible, the algorithm has to use the given laser spectrum which may not be suitable for this specific target pulse train. It is also possible that by using a different fitness function the individual peak heights might be reached better, but probably at the cost of additional unwanted peaks outside of the desired time windows. Using regular M-sequences, 16 pulses would be generated within the pulse train, whereas with the generalized method shown here it is possible to desire specific (and also vanishing) peak heights. Of course the feedback method is not limited to producing pulse trains (which have been generated successfully by other methods [29, 35, 36]), but rather any temporal intensity/phase structure can be selected as a target. Comparing our results with the recently developed technique of direct space-to-time (DST) pulse shaping [36], we note that although with DST shaping pulse trains such as in Fig. 5a could be generated in principle, only fixed-mask shaping of pulse trains with uniform peak heights has been shown to date. Deliberate DST-shaping of the phase has not been reported.

In our case, Fig. 5b indicates that the individual pulses of the pulse train exhibit constant temporal phase. This is in accordance with the expectations because bandwidth-limited laser pulses have been selected as a target, and therefore no additional chirping and temporal pulse broadening is allowed. The temporal phase changes only between the individual pulses. As discussed earlier, however, we did not attempt to fix the phase relationship between these pulses to any specific value.

3 Conclusion

In this paper we described different issues of optimized frequency-domain fs laser pulse shaping. By combining second-harmonic generation (SHG) and an evolutionary algorithm, bandwidth-limited, amplified fs laser pulses of up to 520 μJ pulse energy can be produced without previous knowledge of the input pulse shape. Damage to the optical components is avoided by an appropriate pulse shaper setup.

Feedback-optimized shaping of arbitrary laser pulse shapes is achieved by employing TADPOLE [19], a real-time fs laser pulse characterization method. Experimental limitations as well as optical deficiencies and alignment errors of the pulse shaper are compensated for automatically. Specifically, a local convergence algorithm allows the realization of accurate and reliable spectral chirping, which has many applications in different fields of physics and physical chemistry.

Since TADPOLE yields complete intensity and phase information, optimized tailoring of specific pulse shapes in the time domain is also possible. Employing generalized phase sequences, we used an evolutionary algorithm and direct feedback from the pulse characterization to optimally shape pulse trains with specific peak heights.

It is expected that feedback-optimized pulse shaping will be of considerable value whenever accurate pulse shaping is required.

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