Assessing and managing laser system stability for quantum control experiments

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Stable laser operation, which is essential for quantum control experiments as well as many other phase dependent processes, is investigated with respect to the influence of amplitude and spectral phase noise. Simulations are first performed and an easy to implement experimental method is presented to monitor the amplitude and phase stability of an ultrafast laser system. As an illustration of this stability assessment technique, the data monitoring is used to guide the identification and elimination of fluctuations in the laser amplification process. Through a number of practical alterations of the amplifier configuration, the stability of the laser system was greatly and consistently improved. Fluctuations on different time scales were eliminated, with special emphasis given to maintaining a stable spectral phase. © *2006 American Institute of Physics.*
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I. INTRODUCTION

Achieving quantum control is generally based on adjusting the structure of the spectral phase and amplitude of an ultrashort laser pulse, in order to steer the quantum system dynamics towards a desired state.[1] The basic mechanism behind this process is the manipulation of constructive and destructive interferences amongst the quantum dynamic pathways to optimize the control objective.[2] Since *a priori* design of these laser fields is generally not feasible due to the complexity of the systems and their dynamics, a common approach is to employ a closed-loop search algorithm, such as a genetic algorithm (GA), to guide the optimization of the laser pulse shaper phase and amplitude pixel settings.[3] Many examples of this paradigm are now available including selective molecular fragmentation,[4,5] enhanced high-harmonics generation,[6] and the manipulation of energy transfer in complex biological molecules.[7] Quantum control can be achieved while simultaneously seeking laser fields that produce a good yield and a narrow distribution of such outcomes[8,9] to assure high quality and robust solutions.

Theoretical studies and experimental results show that finding a tailored spectral phase function is often essential for steering the quantum system to a specific goal. This is also evident from the fact that many control experiments can be performed with a phase-only pulse shaper. Even small changes in the phase function $\phi(\omega)$, with ω being the frequency of the laser light, can lead to significant changes in the dynamics of the quantum system under control. Recently, control of the phase of an excited wave packet has been reported,[10] which further underscores the importance of the laser pulse phase for the control of coherent processes. The purpose of this article is to introduce a simple experimental technique to monitor the phase and amplitude stability of a laser system. In addition, we demonstrate how this monitored data can be used to enhance the stability of the amplification process in a particular laser system.

In order to initially investigate the influence of different types of laser fluctuations on the control of quantum systems, computer modeling was performed on the two-photon rubidium atom transition $5S \rightarrow 5D$ and a two-photon diode; both models are part of the LAB2 simulation code.[11] These model systems where chosen because of their relevance to quantum control applications; however the conclusions are likely also valid for many other high-order processes, such as above threshold ionization, high-harmonics generation, attosecond applications, etc., that are extremely sensitive to pulse shape fluctuations. In the case of the Rb transition, a GA was used to find a control field that maximizes the population of the $5D$ state at a certain time. The optimization lead to a phase function that produces a structured pulse train, rather than the featureless bandwidth limited pulse that is optimal for maximizing the signal in the two-photon diode.

For both model systems the respective laser field was then subjected to either pulse energy fluctuations or phase function fluctuations, and the simulations were repeated 2000 times to obtain histograms of the statistical effects on the control outcomes. The results in Fig. 1 show the relative frequency of occurrence on the ordinate and are normalized to the signal without any noise on the abscissa.

The pulse energy in the simulation was drawn from a Gaussian distribution with full width at half maximum (FWHM) of 11.75%. With the optimized phase unchanged, the histograms for the pulse energy, the two-photon diode signal, and the Rb $5D$ state population are shown in Fig. 1(a). The output from the two-photon diode follows the energy fluctuations, but with an increased distribution width due to the nonlinear nature of the process. In the case of the Rb atom excitation, any noise leads to a diminution of the signal, as the outcome is already maximal without noise.

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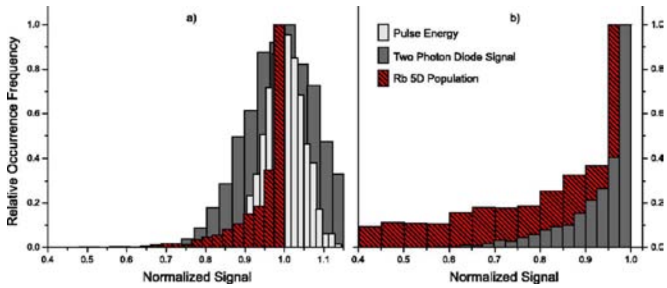

FIG. 1. (Color online) Normalized histograms from simulations with noise in (a) only the pulse energy and (b) only the spectral phase. In both cases the population of the target Rb level decreases from its maximum value when noise of any kind is introduced. In (a) the FWHM of the two-photon diode signal is roughly double the FWHM of the pulse energy, reflective of the second order process, while in (b) any deviation from the optimal phase reduces the diode signal.

The case of phase function fluctuation was treated by defining a model phase function of the form

$$\tilde{\phi}(\omega') = \tilde{\phi}_2 \omega'^2, \tag{1}$$

where ω' is the frequency relative to the center ω_0 (i.e., $\omega' = \omega - \omega_0$). The coefficient $\tilde{\phi}_2$ fluctuates from simulation run to simulation run over a specified Gaussian distribution in order to simulate spectral phase fluctuations. $\tilde{\phi}(\omega')$ was added to the flat phase of the pulse before it enters the pulse shaper, and the resulting pulse phase is the sum of the randomly fluctuating and shaper-set phase. The form in Eq. (1) is a smooth phase distortion over the whole spectrum, with higher terms (i.e., in ω'^3, ω'^4, etc.) left out for simplicity in this model. For the simulation results in Fig. 1(b), the pulse energy was left constant, and the laser spectral phase noise coefficient $\tilde{\phi}_2$ in Eq. (1) fluctuates around zero with a standard deviation of 2000 fs^2 (FWHM of $\tilde{\phi}_2$ 4700 fs^2). For just spectral phase fluctuations, both signals deteriorate as any disturbance of the optimal phase will inherently lead to a nonoptimal reduced signal.

By comparing the histograms in Fig. 1, several conclusions can be drawn. For pulse energy fluctuations, the effect on the two-photon diode signal is to broaden its distribution in Fig. 1(a), as expected. The statistical effect of energy fluctuations on the Rb signal is less than its impact on the diode signal. This behavior may be indicative of energy (amplitude) stability being of moderate importance here and possibly for other quantum control experiments. In contrast, Fig. 1(b) shows that when only the spectral phase is randomly changed, the effect on the control of the Rb atom is much more dramatic, while quantitatively the reduction of the two-photon diode signal is no worse than for energy noise alone in Fig. 1(a). This clearly points towards the varying spectral phase and consequently unstable pulse duration, even at constant pulse energy, that troubles the quantum control simulation.

These simulations illustrate some fundamental issues regarding the use of laser fields to control complex quantum systems. On the positive side, laser energy variations often have a narrow shot-to-shot distribution, thereby reducing their impact on performance. However, for quantum control experiments, the most important stability issue likely arises from spectral phase noise. In a real laser system, instabilities are a combination of energy and phase fluctuations. Therefore, in the remainder of this article, the word stability is used to describe the action of the pulse energy and the spectral phase simultaneously.

Pulse shaping technology is capable of generating structured phase functions with high resolution. However, as the simulations illustrate, if the unshaped pulse from the laser has a fluctuating spectral phase $\tilde{\phi}(\omega')$, it will be passed from the laser through the pulse shaper and into the quantum system, since the pulse shaper can only add a phase function to the unshaped pulse. With significant laser phase noise, the shaped pulses would be of limited use for quantum control experiments that are highly phase sensitive. In some cases it actually may not be possible to find an optimal phase function, as the search algorithm can become constantly confused as to what alterations can improve the control objective. The performance quality of the best pulse shaper can only be as good as the input spectral phase from the laser. While averaging will improve the signal-to-noise ratio, this can be of limited use during learning control experiments using a closed-loop search algorithm, since any additional averaging increases the time needed to run an optimization. Seeking pulses maximally robust to noise may help,[9] but again these special pulse shapes need to be searched for.

While performing a number of quantum control experiments, we encountered both short term noise (i.e., shot-to-shot fluctuations) and slowly varying fluctuations (on the order of several seconds) in the spectral phase of the laser system in our laboratory. The main purpose of this article is to introduce the means to quantitatively identify the presence of laser instabilities. The primary problem we encountered in our particular laser system was slowly varying spectral phase fluctuations on the order of 5–20 s, which is extremely adverse for quantum control experiments employing search algorithms that make judgments on this same time scale. The technique introduced here for identifying such fluctuations was then used to monitor the effects of structural changes made in the laser, and ultimately greatly enhanced the laser stability.

II. EXPERIMENTAL SETUP

The laser employed in this work is a Ti:sapphire femtosecond laser system, consisting of a Spectra Physics Tsunami oscillator and Spitfire amplifier. The repetition rate is 1 kHz and the pulse energy is 1.6 mJ. The output is centered at $\lambda = 797$ nm with $\Delta\lambda \cong 10$ nm bandwidth (FWHM) giving pulses of $\tau \cong 100$ fs pulse length (FWHM).

The output stability of the laser is monitored by splitting off approximately 8% of the laser beam with an uncoated pellicle beam splitter, while the main beam is used for quantum control experiments. Either a one-photon diode or a two-photon process can be utilized to gain information about the stability of the laser; other processes, especially even more nonlinear ones, may be used as well if they can be conveniently monitored. While the signal of the one-photon diode (Si diode with 1.2 ns rise time) depends on the pulse energy, it does not depend on the spectral phase function of the laser

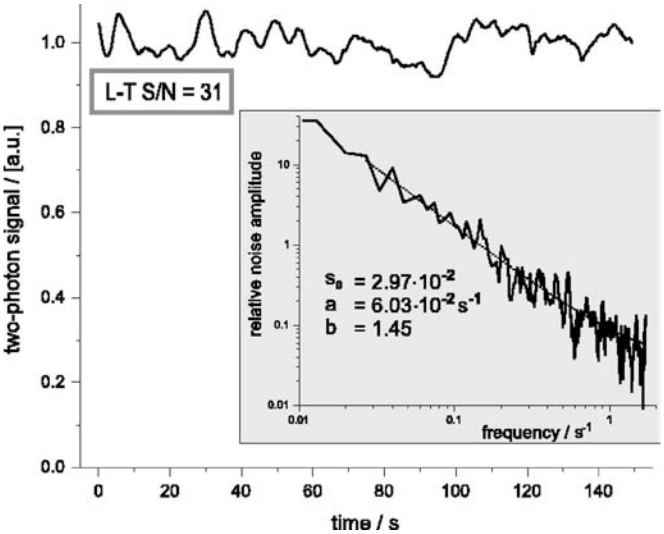

FIG. 2. Typical temporal history of the two-photon diode signal for the laser system operating in an unstable state. The long-term signal-to-noise ratio (L-T S/N) is determined from the time segment of 150 s shown. The inset shows the power spectrum of the two-photon diode noise taken over 30 min. The power spectrum was fitted to $S(f)=s_0+a/f^b$, and the best fit is shown as the dotted line. The parameters s_0, a, and b are used to assess the stability of the laser system together with the L-T S/N ratio.

pulse electric field. For a two-photon process, such as second harmonic generation (SHG) or a two-photon diode, the signal depends on the time dependent intensity of the laser pulse and therefore not only on the pulse energy but also on the phase function $\phi(\omega')$ of the laser field (i.e., ultimately the pulse shape). This behavior of the one- and two-photon diodes is the basis of the laser stability monitoring setup, since the objective is to assure that a stable pulse energy and input phase function of the unshaped pulse enter the pulse shaper.

In the stability monitoring setup, the second order phase sensitive process is two-photon absorption in a GaP diode (1 ns rise time and 140 ns fall time). The diode signal is detected on a shot-to-shot basis with a 1 kHz repetition rate: each millisecond, the diode signal is integrated and digitized with a combination of a boxcar (SR250) and an analog/digital (A/D) converter (SR245). In addition, the signal from a one-photon diode is monitored shot to shot to determine the energy fluctuations. In this fashion it is possible to characterize pulse energy and/or pulse intensity variations.

III. EXPERIMENTAL RESULTS

The shot-to-shot signals are transferred to the computer in blocks of 200 boxcar signals, representing 200 successive laser pulses (i.e., over 0.2 s). These data are then analyzed, where each block of 200 signals is used to get an estimate of the shot-to-shot noise by calculating its standard deviation and mean. The ratio of the mean signal over the standard deviation in any 0.2 s interval is referred to as the *short-term signal-to-noise ratio* (S-T S/N).

In the next step, the mean of each of these short-term blocks is stored continuously in the computer as they are recorded, approximately every 0.3 s, where data transfer and storage take an additional 0.1 s. Figure 2 shows such a typical plot, representing a noisy operating state of the laser system, that is generally not suitable for control experiments. A sequence of 512 of these points is kept, and for each new data point the oldest data point is dropped. The result is a running record of the laser pulse intensity for approximately the last 150 s. In the present laser system, this longer-term picture of the laser stability is primarily a result of the spectral phase (i.e., the one-photon diode signal exhibited fluctuations approximately ten times smaller than from the two-photon diode). In quantum control experiments, when iterative feedback loops are used to find an optimal control field, long-term stability of the phase is essential in order for the learning algorithm to be effective. We choose 150 s as the time window to assess the laser stability, because this is typically on the order of one generation in GA driven optimizations.

The data in Fig. 2 are analyzed in a fashion similar to the procedure for the short-term noise. For each successive window of 512 data points, the mean and standard deviation are calculated, and the ratio of the mean signal over its standard deviation is referred to as the *long-term signal-to-noise ratio* (L-T S/N).

We tested the one-photon signal with the same technique and found it to be generally very stable, with a L-T S/N ratio greater then 300, even if the two-photon signal was very unstable. The one-photon signal is not a measure of the spectral phase, and we use only the two-photon diode data to determine the pulse intensity, which also contains information about pulse energy but primarily depends on the phase function.

For an additional measure of the laser stability, after normalization to unit mean, a fast Fourier transform (FFT) of the signal over a 150 s window is calculated. For the purpose of the stability analysis, we obtain the power spectrum by averaging several of these FFTs (each after another 150 s period) and subsequently squaring the FFT amplitudes. The frequency axis in the present example ranges from 0.0067 Hz (corresponding to the full 150 s segment) out to 1.7 Hz. The power spectrum is fitted to the function $S(f) = s_0+a/f^b$, where s_0 is the frequency independent background noise. The power spectrum and the fit are shown in the inset in Fig. 2. The observed noise power spectrum is likely a complex combination of various sources.

Figure 3 shows the result of the same data analysis as in Fig. 2 after making several modifications to the laser described later in the text. The stabilized laser system clearly shows a significant improvement in its noise characteristics. The parameter a gives the magnitude of noise, and a significant reduction is seen in Fig. 3 over that of Fig. 2. The exponent b shows $\sim 1/f$ noise dependence in the stable case, while the more complex processes operative in the unstable case produce a frequency scaling approximately as $\sim 1/f^{3/2}$. A detailed analysis of the frequency dependence of the signal is beyond the scope of this work.[12–14] However, it is interesting that $1/f$ noise commonly occurs in amplifiers, electronic devices, as well as in many other areas.[15–17]

The software to obtain the data in Figs. 2 and 3 was written in LABVIEW and it enables a continuous monitoring of the stability of the laser. The ratios S-T S/N and L-T S/N as well as the parameters s_0, a, and b of the averaged noise

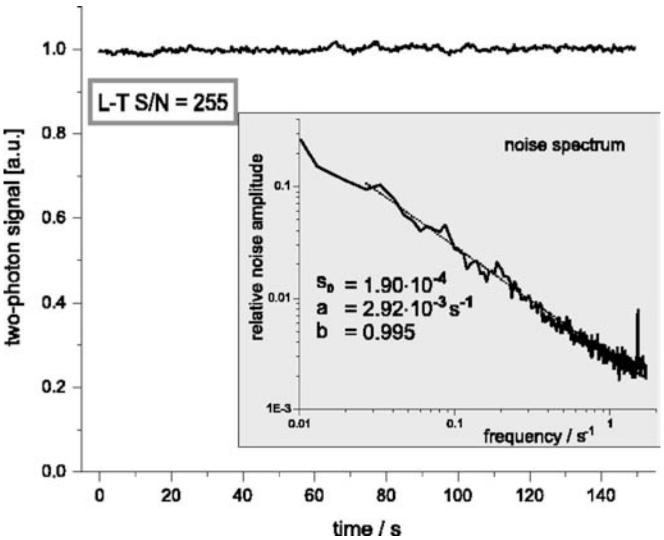

FIG. 3. Typical temporal history of the two-photon diode signal for the laser system operating in a stable state. See the caption of Fig. 2 for further explanations.

power spectrum are continuously displayed. These stability data may be employed in efforts to identify problems in the laser setup that cause instabilities. With the laser used in this work, the data record provided a quantitative means for assessing the impact of various improvements made to the laser. We observed, for example, that the laser could be stable on a short-term shot-to-shot basis but very unstable over the several second time scale.

As an illustration of the value of the stability monitoring technique, we will summarize how it was applied to stabilize the laser. Additionally, the particular findings may be of value to others with similar problems. From our work with the laser system, it appeared that fluctuations in the spectral phase function of the laser pulse in the chirped pulse amplification (CPA) laser system might be introduced in the pulse stretcher and/or the compressor, since in these system components the spectral constituents of the pulse are spatially separated (indicated by the hatched areas in Fig. 4), much like in a pulse shaper. If these spectral constituents pass through air with a fluctuating index of refraction, the phase function $\phi(\omega')$ will fluctuate accordingly. This behavior is well known from the highly unstable operation resulting from opening the enclosure cover of a CPA laser amplifier. However, even relatively slow air currents of slightly different index of refraction can introduce slow variations of the laser pulse spectral phase. In effect, slow air currents in the pulse stretcher or compressor act as a type of chaotic pulse shaper. A similar effect has been observed in the timing jitter of regenerative amplifiers.[18]

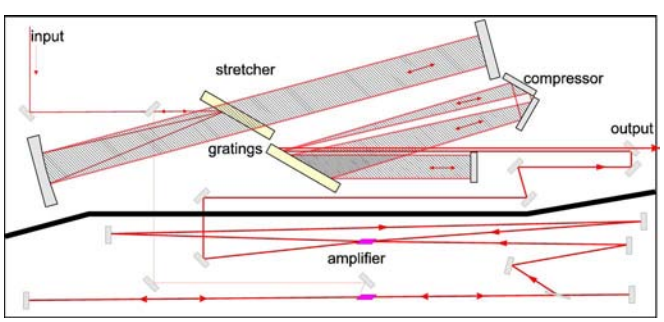

FIG. 4. (Color online) Simplified view of the laser amplifier. The thick black line indicates the added plastic partition wall, which evidentially prevents thermal air currents arising in the amplifier from disturbing the spatially separated spectral components in the stretcher/compressor. The areas where spectral components are spatially separated are indicated by hatching.

For our laser system, we observed small temperature differences of 1–2.5 K between the amplifier and stretcher/compressor regions of the amplifier enclosure, which would likely induce slow flowing air currents. We were able to correlate the time scale of temperature changes with the slowly varying changes in the phase function of the laser pulse, shown in Fig. 2. To address this problem we constructed a plastic partition wall between the amplifier and stretcher/compressor section of the Spitfire amplifier, as indicated by the thick black line in Fig. 4. The partition wall was made to fit tightly, and cracks were sealed using rubber foam. The laser beam passes through small holes in the wall. The laser amplifier and stretcher/compressor regions are also flushed with a slow flow of 4 l/h of dry nitrogen. This was done in order to keep uncontrolled airflows from outside the enclosure away from the laser beams. Importantly, throughout the process of testing these and other trial alterations to the laser, we judged the value of each improvement by using the stability characterization setup and software described above.

The setup also enabled work on improving the shot-to-shot stability by monitoring the short-term signal-to-noise ratio (S-T S/N). However, it was not possible to point out a specific source of this type of noise, as it is likely caused by a variety of factors, including alignment of the seed laser, timing of the input and output trigger of the regenerative amplifier, and spatial overlap of the seed and pump beams in the amplifier. To aid the reduction of the S-T S/N, we also reconfigured the electronics of the laser to use the 80 MHz pulse train of the Tsunami oscillator as a hardware clock for triggering purposes.

Utilizing this strategy of introducing selective alterations in the laser system guided by monitoring the resultant effects on the laser stability with the tools described here, we were able to significantly improve the stability of the laser system. The net result of these adjustments is the much improved noise characteristics shown in Fig. 3. Periodic alignment procedures can now be done not only by optimizing the output energy and pulse length but simultaneously with a focus on laser stability. We believe that routine assessment of laser stability, and, in particular, the presence of a stable input phase function for the pulse shaper, is of prime importance for performing reliable quantum control experiments. Observing only the pulse energy on either short- or long-term time scales is not sufficient for quantum control applications and other phase sensitive processes. The stability characterization tool described above can be used to quantitatively monitor the stability of many laser systems similar to the one employed in this work

IV. DISCUSSION

We have presented an easy to implement technique to assess the stability of both the pulse energy and, generally more importantly for quantum control experiments, the spectral phase function. The technique does not provide the actual phase functions,[19] but rather feedback signals that are sensitive to the laser field fluctuations. Phase stability is essential in many other applications as well, such as an optical parametric oscillator/optical parametric amplifier , parametric inversion, white light continuum generation, etc., and generally many other high-order processes. The technique is simple to implement, because it is not necessary to determine the actual phase function. Rather, the effects on a two-photon process, or possibly another accessible spectral phase dependent physical process, are exploited to establish whether the spectral phase function of the unshaped output from the laser amplifier is changing in an unsatisfactory way over various time scales. Since subsequent shaping of the phase function[20] is a key component in most controlled quantum dynamics experiments, we believe that ensuring a stable input phase is of utmost importance. As an illustration of the utility of this tool, monitoring the laser stability enabled a significant improvement in our laser system. This enhanced stability proved to be of great benefit for both the efficiency and quality of subsequent quantum control experiments. In the future, it would be helpful if quantum control experiments are reported with both the stability data on the input pulses as well as on the controlled dynamics.[9]

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[1] K. Yokoyama *et al.*, J. Chem. Phys. **120**, 9446 (2004).
[2] T. Brixner and G. Gerber, ChemPhysChem **4**, 418 (2003).
[3] R. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992).
[4] R. J. Levis, G. M. Menkir, and H. Rabitz, Science **292**, 709 (2001).
[5] S. Vajda, A. Bartelt, C. Kaposta, T. Leisner, C. Lupulescu, S. Minemoto, P. Rosendo-Francisco, and L. Wöste, Chem. Phys. **267**, 231 (2001).
[6] R. Bartels, M. Murnane, H. Kapteyn, I. Christov, and H. Rabitz, Phys. Rev. A **70**, 043404 (2004).
[7] J. L. Herek, W. Wohlleben, R. J. Cogdell, D. Zeidler, and M. Motzkus, Nature (London) **417**, 533 (2002).
[8] J. Geremia, W. Zhu, and H. Rabitz, J. Chem. Phys. **113**, 10841 (2000).
[9] A. Bartelt, M. Roth, M. Mehendale, and H. Rabitz, Phys. Rev. A **71**, 063806 (2005).
[10] T. Polack, D. Oron, and Y. Silberberg, Chem. Phys. **318**, 163 (2005).
[11] B. Schmidt, M. Hacker, G. Stobrawa, and T. Feurer, http://www.lab2.de, 2000–2006.
[12] E. Milotti, Phys. Rev. E **72**, 056701 (2005).
[13] W. Li, A Bibliography on $1/f$ Noise, http://www.nslij-genetics.org/wli/1fnoise/, 2006.
[14] B. B. Mandelbrot, *Multifractals and* $1/F$ *Noise* (Springer, New York, 1998).
[15] W. Schottky, Phys. Rev. **28**, 74 (1926).
[16] P. Gopikrishnan, V. Plerou, X. Gabaix, and H. E. Stanley, Phys. Rev. E **62**, R4493 (2000).
[17] M. Usher, M. Stemmler, and Z. Olami, Phys. Rev. Lett. **74**, 326 (1995).
[18] T. Miura, K. Kobayashi, K. Takasago, Z. Zhang, D. Torizuka, and F. Kannari, Opt. Lett. **25**, 1795 (2000).
[19] W. Kornelis, J. Biegert, J. W. G. Tisch, M. Nisoli, G. Sansone, C. Vozzi, S. D. Silvestri, and U. Keller, Opt. Lett. **28**, 281 (2003).
[20] A. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).