

C. CORSI¹
M. BELLINI^{2,✉}

Robustness of phase coherence against amplification in a flashlamp-pumped multi-pass femtosecond laser

¹ European Laboratory for Non Linear Spectroscopy (L.E.N.S.), Via N. Carrara, 1, 50019 Sesto Fiorentino (FI), Italy

² Istituto Nazionale di Ottica Applicata, L. go E. Fermi, 6, 50125 Firenze, Italy

Received: 6 June 2003/Revised version: 8 September 2003
Published online: 29 October 2003 • © Springer-Verlag 2003

ABSTRACT We show that the phase of an ultra-short laser pulse undergoing the process of chirped-pulse amplification in a flashlamp-pumped multi-pass configuration is surprisingly well locked to the phase of the original pulse emitted by a mode-locked oscillator. We demonstrate that the relative phase jitter introduced in the amplification remains small (< 0.1 rad r.m.s.) and does not spoil the visibility of fringes obtained in an interferometric experiment. These results indicate that, using a carrier-phase-stabilized seed laser, the generation of phase-controlled, high-intensity laser pulses is possible and relatively straightforward even with the commonly available flashlamp-pumped amplification systems.

PACS 42.65.Re; 42.62.Eh; 42.25.Kb

1 Introduction

The generation of carrier-phase-stabilized laser pulses, i.e. with a phase of the field oscillation controlled with respect to the envelope of the pulse itself, has recently attracted much attention both as a fundamental tool for precision frequency measurements [1, 2] and for its possible implications in the control of phenomena taking place at extremely short time scales [3–5].

Pulses with a controlled carrier-envelope phase (CEP) are at the basis of the broad frequency combs which are revolutionizing the field of high-precision spectroscopy and frequency metrology, and have already allowed the determination of the frequency of atomic and molecular transitions with unsurpassed accuracy [6, 7]. On the other hand, the possibility of manipulating the carrier phase of ultra-short light pulses is expected to give access to a whole range of new phase-dependent phenomena in high-intensity laser–matter interactions, such as strong-field photoionization [4, 5] and high-order harmonic and attosecond pulse generation [8, 9].

Carrier-phase-controlled pulses had however been produced only for low-energy, mode-locked laser oscillators so far, and the transfer of the phase stabilization to the high-energy pulses of an amplified system appeared difficult and

was the subject of strong debate [10]. Only very recently, Baltuška and coworkers [11] demonstrated the generation of intense phase-controlled light pulses and applied them to the control of soft X-ray emission. In their pioneering experiment they made use of a diode-pumped, Q-switched laser with good stability characteristics as the pump for the multi-pass Ti : Sa amplifier.

In this paper we show that the essential phase signatures of an ultra-short, low-energy pulse are not destroyed in the process of multi-pass chirped-pulse amplification (CPA) in a commonly available flashlamp-pumped system. Starting from an unstabilized laser oscillator, we cannot and do not aim to demonstrate any kind of CEP stabilization in our amplified pulses, but we nevertheless provide strong evidence that carrier-phase-stabilized, high-energy pulses can be simply generated by amplification of phase-controlled oscillator pulses. To this purpose, we introduce a simple way of characterizing the phase properties of laser amplifiers, which might be useful for the diagnostics and optimization of systems for the generation of phase-controlled intense ultra-short light pulses.

The essential idea of our work is to compare, in an interferometric (i.e. phase-sensitive) way, a single pulse from a laser oscillator and its replica which has undergone the process of amplification. If a linear correlation technique is used, the observed signal presents an oscillating term with a phase proportional to

$$\Phi = \omega_0 \tau + \varphi_A + \varphi_N, \quad (1)$$

where ω_0 is the central laser frequency, τ is the delay between the pulses, φ_A is the possible phase shift due to the amplification process, and φ_N is any other phase shift not directly connected with the amplification. The simple observation of such an interference on a single-shot basis does not give any indication of the presence of an amplification-induced dephasing, but if the same phase is maintained over many successive laser shots, then it means that the effect of amplification is at least reproducible and can be controlled.

2 Experimental

We performed our experiments with a slightly modified version of a commercial CPA laser system (Femtopower PRO by Femtolasers), consisting of an ultra-short

✉ Fax: +390-55/457-2451, E-mail: bellini@ino.it

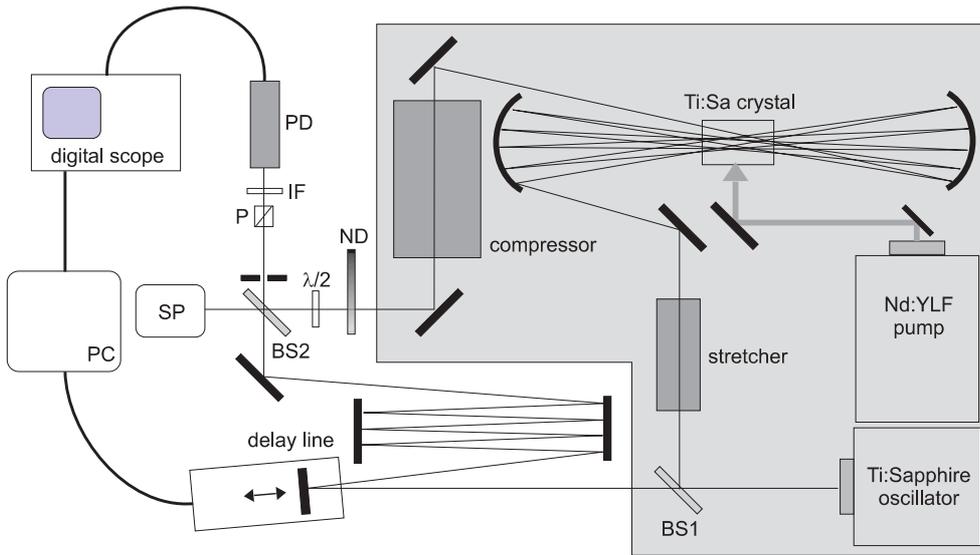


FIGURE 1 Experimental setup. The shaded area delimits the amplified laser source. ND: variable neutral density filter; P: cube polarizer; $\lambda/2$: half-wave plate; IF: narrow-band interference filter; PD: fast photodiode; SP: spectrometer; BS1 and BS2: 50% beam splitters; PC: personal computer. The figure shows the final configuration, where the oscillator pulses are compared with their fully amplified and re-compressed copies; for measurements at different amplification stages, a diverting mirror is placed in the multi-pass path to redirect the pulse out of the system and towards the beam splitter BS2, bypassing the compression stage

(12-fs) mode-locked Ti : sapphire oscillator, whose pulses are first stretched and then amplified in a nine-pass configuration in a second Ti : sapphire crystal pumped by a flashlight-pumped Nd-YLF laser (621D Thales). The re-compression by two pairs of prisms yields 1-kHz repetition rate pulses with a duration of about 25 fs and an energy up to about 1 mJ.

By placing a broadband 50% beam splitter (BS1) before the pulse stretcher, both the oscillator pulses and their amplified versions are emitted from the laser system and their mutual phase coherence can be studied by means of an interferometric setup (see Fig. 1). The experiments were performed by placing a diverting mirror in the path of the amplified pulse at various stages of its multiple passage through the Ti : sapphire amplifier. For each of these positions, measurements were taken at various levels of pump power corresponding to increasing levels of energy amplification.

In order to observe the possible interference effects between a single pulse of the oscillator train and its amplified replica, we took care in measuring the exact optical path through the amplification channel and we compensated it by means of an external optical delay line. The latter was implemented with multiple reflections of the unamplified pulse on table-mounted mirrors, one of which was placed on a motorized translation stage for accurate positioning. The final measurements comparing the original pulse and its fully amplified and re-compressed replica involved a total optical path compensation of about 20 m. Pulses coming from the two paths were superimposed by means of a second 50% beam splitter (BS2), a common polarization component was selected with a cube polarizer, and the amplified pulse was properly attenuated by means of a half-wave plate and variable-density neutral filters.

For all our measurements, the right spatio-temporal matching between the original and the amplified pulses was achieved in several steps. As a first stage, the Q-switched pump was blocked and the pulse-selecting Pockels cell disabled, in order to directly compare two pulse trains, both at 80-MHz repetition rate, the first coming from the oscillator, and the second traveling through the amplification channel. In these conditions it was possible to use a spec-

trometer (SP) to improve the alignment and adjust the proper delay between the pulses in the two channels by observing and optimizing spectral interferences. The long duration (tens of picoseconds) of the chirped pulses passing through the stretcher–amplifier chain allowed us to detect spectral interference fringes in a much wider delay range and vastly facilitated the task of temporally superposing the pulse trains.

Next, a fast photodiode (PD) was used in order to observe the spatially and temporally superposed pulse trains on a digital oscilloscope. Because of the strong chirp in the stretched pulses, it was necessary to use interference filters to restrict the total bandwidth (more than 100 nm) of the pulses to a level where temporal interferences were clearly visible as a periodic modulation of the combined 80-MHz pulse train while varying the relative delay.

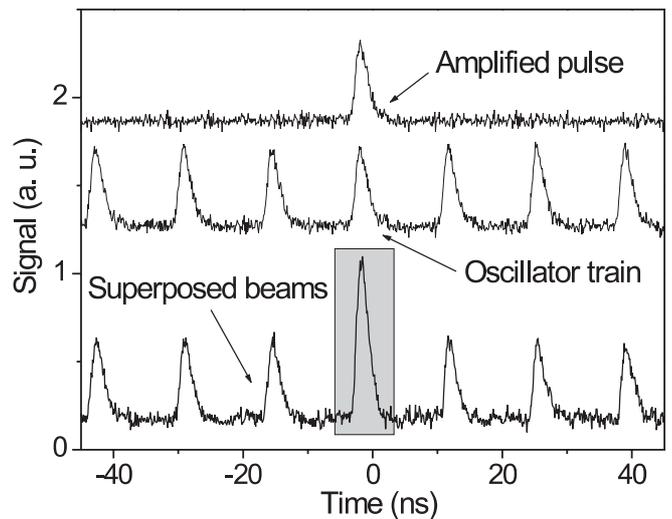


FIGURE 2 Oscilloscope traces of the single amplified pulse at 1-kHz repetition rate (top trace) and of the 80-MHz repetition rate mode-locked pulse train (middle trace) obtained by alternately blocking one of the inputs to the beam splitter BS2. Bottom trace: both inputs to the beam splitter are open and the single amplified pulse is superposed to its parent pulse after spatial, temporal, and intensity matching. Temporal interferences are observed by monitoring the area of the pulse in the shaded box as a function of the optical delay

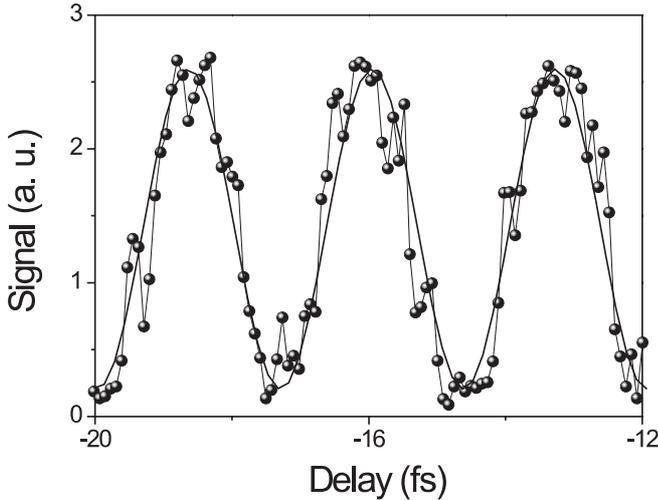


FIGURE 3 Interference signal (*filled circles*) obtained while scanning the delay between the original laser pulse and its amplified replica after nine passes through the Ti : sapphire crystal. Only the central part (close to zero delay) of the whole cross-correlation signal is shown here, together with a fit to a sine function

Finally, the Pockels cell was enabled and the pump laser was unblocked in order to select and amplify a single pulse (at 1-kHz repetition rate) out of the train in the amplifier. Neutral filters were used to balance the energy of this single amplified pulse (top trace of Fig. 2) with that of its twin in the 80-MHz oscillator train (middle trace of Fig. 2). Again, temporal interferences between the original and the amplified pulses were observed as periodic modulations of their combined oscilloscope signal. Data points, each corresponding to a single-shot measurement of the area of the interfering pulse (shaded area in the bottom trace of Fig. 2), were acquired synchronously with the pump-laser pulses at a 500-Hz rate (the maximum allowed by our acquisition system), while finely scanning the optical delay line (see Fig. 3).

3 Results and discussion

In order to single out the possible deleterious effects of pumping (connected to φ_A) from those coming from other unavoidable instabilities in our system (connected to φ_N) on the phase coherence of the amplified pulses, we repeated the measurement at each step in the two conditions of pump on and off. To obtain statistically comparable results, the same kind of synchronization and the same acquisition rate were used for both measurements of the oscillator-oscillator (pump off) and oscillator-amplifier (pump on) first-order cross-correlation signals as a function of the time delay τ .

Figure 4 presents the results corresponding to the measured maximum fringe visibilities, obtained from a fit of the signal to a sinusoidal oscillation at the expected carrier frequency in the region of $\tau \approx 0$ (see Fig. 3), for different amplification factors after two, four, six, eight, and nine passes in the Ti : sapphire amplifier. The last data points correspond to the full final configuration including the nine passes plus the re-compression of the amplified pulse to a duration of about 25 fs. For each step through the amplifier, we also indicate the measured contrast without pumping laser (oscillator-

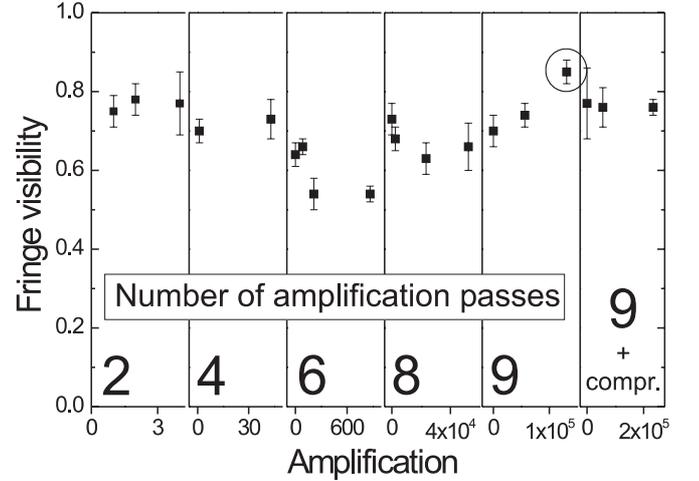


FIGURE 4 Maximum visibility of the interference patterns for different numbers of passes through the amplifier and for different values of the overall amplification factor. The first point in each sub-set is obtained with the pump switched off. The point in the *circle* is extracted from the data of Fig. 3

oscillator correlation, corresponding to an amplification factor of 1). It is evident that, apart from a somewhat erratic behavior of the contrast, probably connected to slightly different alignment conditions in the different measurements, there is no substantial worsening in the visibility of the fringes when amplification is present, even at the maximum pump power, corresponding, for the last data point, to a pump pulse energy of about 5.5 mJ and to an amplified pulse energy of about 0.6 mJ.

As a further test of the stability of the relative phase, we also took recordings of the central zone of the interference pattern while keeping the delay fixed. We observed fast instabilities as well as slower drifts in the relative phase, but in neither case did we find an apparently higher noise when the original pulses were made to interfere with their amplified replicas, with respect to interference with their simply delayed versions, thus suggesting that environmental instabilities (and not pump-related phase factors) were the main sources of phase noise. In some cases fringes were observed to remain stable for times as long as 5 s, indicating that the same phase relationship between the original and the amplified pulses can be maintained for several thousands of shots.

Note that, if instead of exactly compensating the total optical path difference between the original and the amplified pulses, we shortened the external delay line to the point of comparing the amplified pulse with the one corresponding to the fifth after its parent pulse in the train, we still observed some interferences, but the stability of the fringe pattern got much worse. The same unstable behavior was observed both when the pump laser was on and off. Although the optical path length outside the laser had been drastically reduced in this case, which should have led to lower environmental instabilities, the higher fluctuations are here caused by the fact that our laser oscillator lacks any form of phase stabilization. The relative carrier phase within the pulse envelope is not stabilized from pulse to pulse and leads to a rapid fluctuation of the interference pattern measured between distinct pulses.

The task of determining in a more quantitative way the contribution of the amplification to the phase noise (by subtracting the environmental phase jitter, which does not depend on the pump) is significantly complicated by the presence of a non-negligible and varying amount of amplitude noise. We isolated such a contribution in regions where it is the only noise source (close to maxima or minima of the fringe pattern or for long delays, where the two pulses no longer interfere), and the approximate amount of phase noise was then extracted by analyzing the regions of fringe zero crossing (where the dependence on the phase is almost linear), with the assumption of uncorrelated amplitude and phase fluctuations. The r.m.s. phase jitter in the case of full amplification and recompression was about 0.4 rad and did not change, within the accuracy of our measurement procedure, while switching on and off the pump laser. We can then prudently assume 0.1 rad as the upper limit for the total r.m.s. phase jitter introduced by our multi-pass amplification system.

An alternative procedure, based on the integration of the noise spectra of signals obtained by scanning the delay at fixed speed, gave similar results, revealing substantially flat noise spectra around the imposed carrier frequency of fringe modulation (≈ 20 Hz), with a few small peaks, probably connected to mechanical resonances. Note that both approaches consider the same frequency interval, between a few tens of Hz and 250 Hz. While the upper limit is fixed by the repetition rate of the laser and of the acquisition system, the lower limit only excludes very low frequencies where the amplification-induced phase noise can be kept small with a simple thermal stabilization of the amplifier. Again, the lack of measurable variations in the integrated noise spectra when the pump laser was switched on is a further confirmation that the 0.1 rad value is a confident upper-limit estimation for the r.m.s. phase jitter due to amplification.

Our measurements were performed with a pump laser having rather poor stability characteristics. The pulse to pulse energy stability of our flashlamp-pumped laser was measured to be about 1%–2%. The effects of such energy variations on the phase of the amplified pulse via cross-phase modulation are however expected to be rather small. Considering a total path of about 10 cm in the amplifying medium with a non-linear refractive index $n_2 \approx 3 \times 10^{-16}$ cm²/W and a peak pump intensity of about 5×10^9 W/cm², then the phase jitter due to amplitude-to-phase noise conversion remains less than 0.1 rad as long as the energy stability is better than 10%. Possible shot-to-shot variations in the intensity of the amplified pulse itself are not expected to cause significant phase jitter due to self-phase modulation. Although in the last pass through the amplifier the pulse energy reaches the millijoule level, the pulses are not too short (still about 10-ps long) and the beam is not focused too tightly, so that peak intensities in the medium are limited. Indeed, the integrated peak non-linear phase shift imposed on the pulses by the optical Kerr effect in the amplifier crystal (B integral) is estimated to be < 0.5 , and the typical pulse intensity fluctuations of our system ($< 3\%$) should thus have a negligible impact on the phase noise. Another proposed source of phase noise in the amplified pulse, connected with beam-pointing effects in a grating-based stretcher [10], is not of concern here, since the stretching effect is obtained by propagation in bulk heavy-flint glass.

A solid-state-pumped amplified laser, like the one used in [11], would allow a better energy stability and would help to further limit the phase jitter in the case of demanding applications. Both the pulse to pulse energy stability of their pump laser and of their amplified pulses are reported to have a standard deviation of less than 1%, which translates into a corresponding reduction (by a factor of 2–3) in the expected induced phase noise via cross- and self-phase modulation when compared with the typical situation of a flashlamp-pumped system. A more direct comparison of the amplification-induced phase jitter with [11] is made difficult by the lack of a quantitative statement about such phase noise by the authors. They measure a 50 mrad r.m.s. jitter introduced by the hollow-fibre-chirped-mirror pulse compressor, but only a qualitative description is given of the (low) phase noise introduced by their multi-pass amplifier.

4 Conclusions

We have found that the process of chirped-pulse amplification in a flashlamp-pumped multi-pass amplifier does not introduce substantial phase jitter on the ultra-short laser pulses emitted by a mode-locked laser oscillator. Comparing the initial laser pulse with its amplified replica by means of a phase-sensitive interferometric setup, we have experimentally demonstrated the robustness of their mutual phase coherence against multiple passes in the amplifying medium and up to very high gain factors. These results make us confident that, with the use of a properly carrier-phase-stabilized seeding laser, ultra-short and high-intensity pulses with precisely controlled phase can be generated even with the now commonly available flashlamp-pumped systems. The broad availability of few-cycle intense pulses with such characteristics will open the way to entirely new fields of research in high-intensity laser–matter interactions and attosecond physics.

ACKNOWLEDGEMENTS We gratefully thank T.W. Hänsch for many inspiring discussions and for his constant support. This work was also partly supported by EEC contract HPRI-CT1999-00111.

REFERENCES

- 1 J. Reichert, R. Holzwarth, T. Udem, T.W. Hänsch: *Opt. Commun.* **172**, 59 (1999)
- 2 R. Holzwarth, T. Udem, T.W. Hänsch, J.C. Knight, W.J. Wadsworth, P.S.J. Russell: *Phys. Rev. Lett.* **85**, 2264 (2000)
- 3 T. Brabec, F. Krausz: *Rev. Mod. Phys.* **72**, 545 (1999)
- 4 G.G. Paulus, F. Grasbon, H. Walther, P. Villoriesi, M. Nisoli, S. Stagira, E. Priori, S. De Silvestri: *Nature (Lond.)* **414**, 182 (2001)
- 5 D.B. Milošević, G.G. Paulus, W. Becker: *Phys. Rev. Lett.* **89**, 153001 (2002)
- 6 J. Reichert, M. Niering, R. Holzwarth, M. Weitz, T. Udem, T.W. Hänsch: *Phys. Rev. Lett.* **84**, 3232 (2000)
- 7 M. Niering, R. Holzwarth, J. Reichert, P. Pokasov, M. Weitz, T.W. Hänsch, P. Lemonde, G. Santarelli, M. Abgrall, P. Laurent, C. Salomon, A. Clairon: *Phys. Rev. Lett.* **84**, 5496 (2000)
- 8 A. Apolonski, A. Poppe, G. Tempea, C. Spielmann, T. Udem, R. Holzwarth, T.W. Hänsch, F. Krausz: *Phys. Rev. Lett.* **85**, 740 (2000)
- 9 M. Hentschel, R. Kienberger, C. Spielmann, G.A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, F. Krausz: *Nature (Lond.)* **414**, 509 (2001)
- 10 F.W. Helbing, G. Steinmeyer, U. Keller, R.S. Windeler, J. Stenger, H.R. Telle: *Opt. Lett.* **27**, 194 (2002)
- 11 A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V.S. Yakovlev, A. Scrinzi, T.W. Hänsch, F. Krausz: *Nature (Lond.)* **421**, 611 (2003)