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Multi-delay, phase coherent pulse pair generation for precision Ramsey-frequency comb spectroscopy

J. Morgenweg* and K. S. E. Eikema
LaserLaB, Department of Physics and Astronomy, VU University, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands
*jonas@few.vu.nl

Abstract: We demonstrate the generation of phase-stable mJ-pulse pairs at programmable inter-pulse delays up to hundreds of nanoseconds. A detailed investigation of potential sources for phase shifts during the parametric amplification of the selected pulses from a Ti:Sapphire frequency comb is presented, both numerically and experimentally. It is shown that within the statistical error of the phase measurement of 10 mrad, there is no dependence of the differential phase shift over the investigated inter-pulse delay range of more than 300 ns. In combination with nonlinear upconversion of the amplified pulses, the presented system will potentially enable short wavelength (<100 nm), multi-transition Ramsey-frequency comb spectroscopy at the kHz-level.

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1. Introduction

The realization of frequency combs (FC) based on mode-locked oscillators has enabled the production of phase-coherent optical pulse trains [1, 2] and the direct measurement of optical frequencies, leading to various applications such as atto-science [3], quantum control [4] and precision metrology [5]. While for many purposes the output power of an unamplified FC is sufficient, there are many applications which require a higher peak power than an oscillator can deliver. One application in particular is nonlinear upconversion of FCs (which typically operate in the near-infrared) to the extreme ultraviolet (<100 nm) for ultra-high precision spectroscopy. In this wavelength region simple atomic and molecular systems, such as He, He⁺ or H₂, have their principal transitions which can be used for highly-accurate tests of the theory of quantum electrodynamics [6–8].

One experimental approach to increase the FC pulse intensity uses an enhancement cavity for efficient high harmonic generation [9, 10], and FC generation in the extreme ultraviolet [11]. As an alternative route, we demonstrated that pulse pairs from a Ti:Sapphire FC can be amplified while maintaining phase coherence; the amplified pulses can then be used to perform high-precision Ramsey-like FC spectroscopy [12, 13]. More recently this principle was employed with an improved system based on an optical parametric chirped-pulse amplifier (OPCPA) and high-harmonic generation, which resulted in MHz-level FC-spectroscopy in Helium at 51 nm [6].

The parametric amplification of two FC pulses requires a pair of high-energy pump pulses,
which in [6] were created by splitting a single pump pulse via an optical delay line. This method introduced two main limitations. First, the physical delay line limited the maximum possible temporal delay to about ten nanoseconds. This in turn limited the resolution of the Ramsey-like spectroscopy [14], and changes of the pulse delay at the nanosecond level required a physical change of the delay line and realignment of the amplifier system. Second, due to the additional optical path for the delayed second pump pulse, notable wavefront deviations were introduced, which resulted in phase shifts of the amplified FC pulses in the OPCPA. These phase shifts had to be monitored constantly with high accuracy (≈ 20 mrad) in order to correct the spectroscopy signal.

In order to lift both these limitations a new pump front-end system based on fast modulators and ultra-high gain Nd:YVO₄ grazing incidence amplifier has been developed [15, 16]. In conjunction with a Nd:YAG post-amplifier and second harmonic generation, 532 nm pulse pairs at the 100 mJ-level and programmable delays up to the microsecond range can be produced. These pump pulse pairs then amplify two pulses from a synchronously locked Ti:Sapphire FC in an OPCPA, similar to the one used in [6].

The upgraded system opens the new possibility to obtain Ramsey-like signals at different and much longer time delays (at multiples of the round-trip time of the oscillators) without any physical change in the setup. If the phase influence of the OPCPA system is constant at different time delays, then this OPCPA phase shift can be eliminated completely by comparing Ramsey signals at different time delays. This is a fundamental difference to traditional Ramsey-spectroscopy, where the absolute phase shift determines potential frequency errors [17].

It is, however, of vital importance that a potential absolute phase shift between amplified FC pulse pairs does not depend on the inter-pulse delay $T$. A phase shift difference $\Delta \phi$ between pulse pairs with inter-pulse delays that differ by $\Delta T$ would cause a systematic frequency error of

$$\Delta f = \frac{\Delta \phi}{2\pi \Delta T}.$$  (1)

For example, a phase shift difference of $\Delta \phi = 5$ mrad for $\Delta T = 8$ ns translates into a 100 kHz frequency error.

The systematic investigation of phase shifts caused by the OPCPA system, and in particular their dependence on the inter-pulse delays, constitute the main part of this paper. First, the investigated experimental setup is outlined in Sec. 2. In order to estimate the phase sensitivity of the amplified FC pulses to experimental parameters like pump pulse intensity and phasematching angle, simulations of the narrow-band OPCPA were carried out which are discussed in Sec. 3. The outcome of the theoretical considerations then provide the road-map for the experimental investigation of the pump pulse pairs for the OPCPA (Sec. 4). Finally, in Sec. 5 the results of the actual phase shift measurements of the amplified FC pulses are presented and discussed.

2. Experimental setup

Figure 1 shows a schematic of the experimental setup. A Nd:YVO₄ pump front-end, quasi-continuously pumped at 28 Hz, delivers picosecond 1064 nm pulse pairs at the mJ-level and is described in detail in [15, 16]. The seed pulses of the pump pulse amplifier are picked via fast modulators from a home-built, passively mode-locked Nd:YVO₄ oscillator. Two ultra-high gain grazing-incidence Nd:YVO₄ amplifier slabs are then employed to boost the pulse energy of the picked pulse pairs to the mJ-level. As opposed to creating the pump pulse pairs via an optical delay line as employed in [6], all pulses travel exactly the same optical path in order to reduce phase effects in the OPCPA due to wavefront differences of the pump pulses. In addition, the inter-pulse delay $T$ within one pump pulse pair can be changed by multiples of the cavity
round-trip time of the pump oscillator ($T_0 = 8$ ns) via a programmable delay generator.

Fig. 1. Schematic of the experimental setup, including a Pockels cell pulse picker (PC), second harmonic generation (SHG) and the optical parametric chirped-pulse amplifier (OPCPA). A reference fraction of the non-amplified frequency-comb pulses is compared with the amplified pulses via spectral interference in the phase-measurement setup. The programmable inter-pulse delay can be changed in steps of the cavity round-trip time ($T_0 = 8$ ns, $n$ is an integer number).

A flash-lamp pumped Nd:YAG post-amplifier further boosts the pulse energy to 140 mJ, and after frequency doubling two 532 nm pulses at the 100 mJ-level are available. In between pre- and post-amplifier, a Pockels-cell pulse picker can be used for additional pulse shaping by adjusting the intermediate energies of the amplified pulses.

The repetition rate of the pump oscillator is locked to a stable reference frequency, synchronously with a home-built Ti:Sapphire FC, which functions as the seed laser for the OPCPA. Because of the common repetition rate, the FC pulses are then selectively amplified according to the programmable inter-pulse delay of the pump pulses. The three-stage, narrowband OPCPA system consists of an unsaturated first pass followed by two saturated passes and produces amplified Ti:Sapphire FC pulses of more than 5 millijoule pulse energy [6].

The pulse-to-pulse phase shift of the FC seed pulses from the oscillator is known from the f-2f feedback loop of the FC. In order to measure additional phase shifts induced by the OPCPA, the phases of the two amplified FC pulses were compared with the technique of linear spectral interferometry, using 25% of the FC seed power split off before the OPCPA as a reference [18, 19]. By comparing the spectral interference patterns of the two FC pulses, the differential spectral phase induced by the OPCPA can then be determined via a Fourier-transform method [20].

The OPCPA phase shift of the amplified FC pulses crucially depends on the pump pulses used for the parametric amplification. Therefore potential pump pulse differences were investigated by optically gating either the first or the second pulse of the frequency-doubled pump pulse pair. A Shack-Hartmann wavefront sensor and a CCD camera were then used to record the differential wavefronts and different intensity profiles, respectively.

3. **Numerical simulations of a narrow-band OPCPA system**

Prior to quantitatively analyzing the amount of phase shift on the (FC) seed pulses in the OPCPA and the possible causes, it is instructive to investigate the influence of certain experimental parameters via numerical simulations. Therefore a split-step method was used to numerically solve the coupled equations for parametric amplification, similar as described in [21]. In order to extract only the nonlinear phase caused by the parametric interaction, the linear phase originating from material dispersion was subtracted from the presented simulation results.
The parameters used in the simulation represent a typical operational condition of our three-stage OPCPA system. The signal (seed) beam was modelled as an initially 10 fs transform-limited Gaussian pulse, clipped to 6 nm bandwidth and centered around 780 nm. This pulse is stretched to about 10 picosecond duration by applying 690000 fs² of group delay dispersion; the seed intensity was taken as 450 W/cm². The pump pulses were assumed to be 58 ps Gaussian-shaped pulses with an intensity equal to 5.5 GW/cm² (40% less in the third stage).

In Fig. 2(a) the spectral output intensity and phase is presented for four different pump intensities in steps of 0.1 GW/cm². The characteristic peaks of the spectral intensity at the edges of the spectra are due to a combination of saturation effects in the parametric amplification process and the hard spectral clipping of the seed spectrum. It can be seen that although the intensity undergoes notable changes, the effect on the spectral phase is much less pronounced. Only in the spectral wing regions, the signal phases differ by more than several tens of milliradians.

It should be noted that the spectral phases in the left column plots of Fig. 2 are simply the sum of the individually acquired spectral phases in the different OPCPA passes. The contributions from the individual passes are shown in the right column of the figure, where the spectral phase changes (relative to the obtained spectral phase for the lowest simulated pump intensity of $I_p = 5.3$ GW/cm²) have been plotted.

\[ \Delta \phi_s(L) = \Delta k \int_0^L \frac{f(z)}{f(z) + \gamma z} dz, \]  

Fig. 2. Simulation results for the investigated narrow-band OPCPA system. (a): Resulting changes of the amplified spectra and the nonlinear phase due to variations in pump intensity. (b): Detuning the phase-matching angle from $\phi_{PM,0} = 2.30000^\circ$ while keeping the pump intensity fixed at $I_p = 5.5$ GW/cm². In the right column of the figure the corresponding differential phase contributions from the individual OPCPA passes are shown.

It is apparent from Fig. 2(a) that the applied changes in pump intensity hardly have any effect on the spectral phase after the unsaturated first pass (note the different y-axis scales). This effect can be understood by looking at the analytical expression for the change in signal phase after parametric amplification in a crystal of length $L$ [22]:

\[ \Delta \phi_s(L) = \Delta k \int_0^L \frac{f(z)}{f(z) + \gamma z} dz, \]
which depends on the detuning $\Delta k$ from the ideal phase-matching condition, the spatially dependent pump depletion $f = 1 - I_p(z)/I_p(z=0)$ and the initial ratio of pump and seed intensity $\gamma_s^2 = \lambda_s I_s(z=0)/\lambda_p I_p(z=0)$; $\lambda_s$, $I_s$ and $\lambda_p$, $I_p$ are the intensities and center wavelengths of signal and pump, respectively. In the unsaturated regime $\gamma_s^2 << f$, hence the integrand in Eq. 2 is close to unity and changes in the pump intensity have little effect on the amplified signal phase.

The influence of the initial signal (seed) intensity on the amplified spectra was found to be even significantly smaller than the influence of changes in pump intensity. A change of 5% in seed intensity caused a phase shift difference of $\approx 1$ mrad, hence phase shifts and phase jitter due to seed intensity fluctuations from the Ti:Sapphire FC are typically not of a concern.

Equation 2 furthermore suggests a linear dependence of the induced phase shift with changes of the phase mismatch $\Delta k$, therefore simulations were performed for different phase matching detunings, relative to $\phi_{PM,0} = 2.3000^\circ$. As apparent from Fig. 2(b), already changes on the order of a few tens of microradians can cause significant (hundreds of milliradians) phase shifts over the entire spectrum, although the spectral intensity remains basically unaltered. Note that in the simulations the phase mismatch for the third pass was reduced by a factor five with respect to the first two passes, according to the five times larger pump beam and hence five times smaller wavefront deviations expected in the third pass of the OPCPA. This explains the smaller relative phase shifts in the third pass as seen from the individual contribution plots (right column of Fig. 2).

Not included in the above simulations are the effects of self-phase modulation (SPM) and cross-phase modulation (XPM) between signal, pump and idler beam. However, these phase shifts can be estimated by calculating the B-integral (type 1 phase-matching) in a crystal of length $L$ [23]:

$$B_{tot} = B_{ss} + B_{ps} + B_{is} = \frac{2\pi}{\lambda_s} \int_0^L n_2 \left( I_s(z) + \frac{2}{3} I_p(z) + 2I_i(z) \right) dz.$$  (3)

In order to investigate their relative impact, the total value of the B-integral ($B_{tot}$) was split up into the contributions from the signal beam itself due to SPM ($B_{ss}$) and from the coupling of the pump and idler intensities with the signal beam via XPM ($B_{ps}$ and $B_{is}$, respectively). Assuming a nonlinear coefficient of $n_2 = 4 \times 10^{-16}$ cm$^2$/W for BBO around the signal wavelength of 780 nm [24], this resulted in the phase shifts depicted in Fig. 3.

![Fig. 3. Calculated B-integral values of the three stage OPCPA for different pump intensities. Shown are the total value ($B_{tot}$) and the individual contributions from SPM of the signal beam ($B_{ss}$) and XPM between pump and signal ($B_{ps}$) and idler and signal beams ($B_{is}$).](image)

It can be seen that most of the phase shift is caused by XPM between the signal and pump beam, where a change of 5% in pump intensity causes an additional phase shift of about 10 mrad of the amplified signal phase. Similar relative changes of the seeding intensity result in a two orders of magnitude lower phase shift and can therefore be neglected.
4. Analysis of the pump pulse pair

As mentioned in Sec. 2, one of the main reasons for the development of the new pump-frontend was to assure that all amplified pulses travel exactly the same optical path, which enables the production of pump pulse pairs with equal wavefronts. However, the high saturation level of the ultra-high gain pre-amplifier (net gain >70 dB) introduces the issue of temporal and spatial gain shaping. In this section, we discuss the influence of these shaping mechanisms, as well as the direct measurement of the wavefront deviations within one pump pulse pair.

4.1. Temporal gain shaping

Because the first seed pulse takes out a significant part of the stored energy in the amplifier crystals, the gain of the amplifier becomes a function of time. The instantaneous gain $G(t)$ can be modelled as

$$G(t) = \frac{G_0}{G_0 - (G_0 - 1)\exp\left[-\frac{F_{in}(t)}{F_{sat}}\right]}$$

and depends on the undepleted gain factor $G_0$, the saturation fluence of the amplifier crystal $F_{sat}$ and the integrated input intensity $I_{in}$ from a starting time $t_0$ up to the time $t$,

$$F_{in}(t) = \int_{t_0}^{t} I_{in}(t')dt'.$$

Figure 4 shows the calculated pulse shapes after our three stage Nd:YVO₄ pre-amplifier ($F_{sat} = 170 \text{ mJ/cm}^2$ and a total undepleted gain factor of $G_{0,\text{tot}} = 90$ dB) and after the double-pass flash-lamp pumped Nd:YAG post-amplifier ($F_{sat} = 650 \text{ mJ/cm}^2$ and $G_{0,\text{tot}} = 25$ dB) for seeding energies at the pJ-level for the first stage. For simplification, a flat-top spatial distribution was assumed for the laser pulses and losses in the amplifier were neglected.

![Fig. 4. Calculated amplified pulse shapes for different seeding ratios (first pulse : second pulse) of the two pump pulses to obtain equal pulses after the post-amplifier. Each row shows the pulse shapes upon exiting the pre-amplifier (left column) and post-amplifier (right column). In c), the pulse intensity of the first pulse was reduced by 47% between pre- and post-amplifier, which in the experiment is performed by a Pockels cell pulse picker.](image-url)
The effect of gain saturation is most apparent in the case of equal seeding energy into the pre-amplifier as seen in Fig. 4(a), where the amplified pulses differ greatly in intensity and shape. The most straightforward way to achieve equal peak intensities is to adjust the seeding energy (Fig. 4(b)), but the different gain dynamics result in a ≈9 ps shift of the intensity maximum of the pulse. Since the repetition rate of the pump oscillator is synchronized to the Ti:Sapphire FC, this shift would cause the FC pulses to see different parts of the pump pulses in the OPCPA, thus experiencing different amplification.

However, the simulations indicate that this issue can be addressed by cutting away about half the energy of the first pulse after the pre-amplifier as seen in Fig. 4(c). This additional pulse shaping, in combination with adjustment of the seeding energies, allows the production of two almost identical amplified pulses despite the strong gain saturation. In the experimental setup, a fast Pockels cell pulse picker is employed to reduce the pulse energy of the first pulse after the pre-amplifier (see Fig. 1).

4.2. Spatial gain shaping

The seeding pulses for the pre-amplifier have a Gaussian-like spatial intensity distribution, hence the saturation of the amplifier will also cause spatial gain shaping. This means that the spatial profile will be distorted after amplification. Furthermore, since the first pulse takes out a significant part of the stored energy in the amplifier, the second pulse will be distorted differently due to a different spatial distribution of the remaining stored energy. The result can be seen in Fig. 5, where beam profiles of the first and second pulse after the post-amplifier (with different time delays of 40 ns and 400 ns) are shown.

![Fig. 5. Amplified beam profiles of the first pulse and two second pulses with different time delays of 40 ns and 400 ns, respectively.](image)

It is apparent that although the overall shape of the beam profiles are very similar (as expected since the pulses travel the same optical path in the amplifier), the intensity profiles of first and second pulses can still differ significantly over the beam. Different pump intensities influence the phase of the amplified FC pulses as described in Sec. 3, thus the spatial gain shaping will cause a spatially dependent amplifier phase shift. However, Fig. 5 also shows that the spatial differences (and hence the induced spatially dependent phase shifts) basically remain constant with different time delays, because the gain distribution after the first pulse hardly changes on a timescale less than one microsecond.

4.3. Wavefront differences

While relevant differences in spatial intensity distributions on the percent-level can be measured straightforwardly with a CCD-camera, it is experimentally challenging to measure wavefront deviations on the microradian-level. A Shack-Hartmann wavefront sensor was used in order to
establish relative wavefront deviations between the first and a second pulse at a 40 ns inter-pulse delay. Figure 6 shows the obtained wavefront tilts in horizontal and vertical direction across the beam as a result of an average over 40 single-shot acquisitions. It is apparent that the differential wavefront tilt is zero within the statistical measurement error of about 50 μrad. However, the measurement accuracy is not sufficient to detect wavefront tilts below tens of microradians, which can still influence the phase of the amplified FC pulses (see Sec. 3). In the next section the phase shift is therefore quantified based on direct measurements using spectral interferometry.

Fig. 6. Relative horizontal and vertical wavefront tilts between the first and a second pump pulse at 40 ns inter-pulse delay, based on an average of 40 acquisitions.

5. Phase measurements of the amplified frequency comb pulses

5.1. Phase shifts at different pulse delays

The ultimate test of the system is a direct measurement of the differential phase shift of the amplified FC comb pulse pairs after the OPCPA. Therefore the spectral phase-measurement setup (see Sec. 2) was used to determine this phase shift and its dependence on the inter-pulse delay.

Figure 7 shows two of these measurements performed on different days and states of alignment of the OPCPA. During the phase-measurement, the inter-pulse delay of the pump pulses was changed in steps of 80 ns and Fig. 7(b) shows the average differential phase shifts per pulse delay; the statistical error of the averaged value of ≈10 mrad is mainly due to noise from the phase measurement procedure itself. It can be seen that independent of the absolute differential phase shift, which is alignment-dependent and can be as big as a few hundred milliradians, the differential phase shift effectively remains the same for different inter-pulse delays. Together with the numerical simulations discussed in Sec. 3, this indicates that the average wavefront tilt between the first and the second pump pulse remains equal on a microradian level, independent on the inter-pulse delay. Furthermore, if one assumes a linear relation between differential phase shift and inter-pulse delay, the statistical error of the phase measurement together with the maximum investigated pulse delay can be used to obtain an upper bound on the inter-pulse de-
lay dependent phase shift. According to Eq. 1, this potential systematic phase shift of \( \approx 10 \) mrad would correspond to a frequency error of less than 5 kHz at the fundamental laser frequency.

However, if the same 10 mrad can be attained over a longer delay, then the corresponding error reduces even further with this delay. In order to investigate significantly longer pulse delays than the 336 ns presented in this paper, a rebuild of the phase measurement setup is required, incorporating an additional Pockels cell to suppress the intermediate reference pulses. While not producing a spectral interference pattern, these pulses still contribute to the measurement noise by adding a constant background level to the interference signal and thus limited the investigated delays to a few hundred nanoseconds.

![Fig. 7. (a): Differential phase shift measurements between two amplified FC comb pulses, for two states of alignment of the OPCPA system (Set 1 and Set 2, respectively). During one measurements the inter-pulse delay was changed in steps of 80 ns. (b): Average phase shifts for each inter-pulse delay of these two measurement sets together with their statistical uncertainties.](image)

5.2. Phase shift scaling with pulse intensity ratio

From the simulations discussed in Sec. 3 it is clear that for a constant differential phase shift of the amplified FC pulses, a stable intensity ratio of the pump pulse pair is essential. Therefore the pump pulse intensity ratio was changed while measuring the phase of the FC pulses for a constant inter-pulse delay. The resulting phase shifts for a certain alignment state of the OPCPA are shown in Fig. 8, together with a linear fit of the experimental data points. In general, the linear scaling coefficient depends on the amplifier alignment, but is typically \( \lesssim 1 \) mrad per 1% pulse energy difference. Hence as long as the pump pulse intensity ratio is kept stable within a few percent, the resulting phase jitter can be kept below 10 mrad rms. To avoid drifts which would cause systematic phase shifts, the Pockels cell unit in between pre- and post-amplifier (see Sec. 2) is used in a simple feedback loop to stabilize the pump pulse energy ratio.

The experimentally determined scaling factor is typically about half the value that was expected from the numerical simulations in Sec. 3. One possible reason is that in the simulation the phase shifts after each pass were simply added, while in reality they can mutually cancel each other over the three amplifier stages in the OPCPA.
5.3. Spatial dependence of the phase shift

In order to examine the spatial dependence of the measured phase shifts, a pinhole was used to select different spatial parts of the amplified FC pulses (see Fig. 9, inset). For each subsection a short phase-measurement was then carried out, while switching the inter-pulse delay. As can be seen from Fig. 9, the phase shift can differ by \( \approx 100 \text{ mrad} \) for different parts of the amplified FC pulses. These deviations are mainly attributed to the different spatial intensity profiles of the first and second pump pulse (see Fig. 5). In addition, nonlinear effects like SPM can already have an influence in the last stages of pre- and post amplifier of the pump laser. This can potentially cause slight (few \( \mu \text{rad} \)) wavefront differences between the first and second pump pulse due to different intensity profiles, which in turn could lead to phase shifts in the amplified FC pulses.

However, when the amplified pulses are used for a spectroscopic experiment, typically only the average spatial phase shift matters, so that the measured spatial variations will at most cause a slight reduction in contrast of the measured Ramsey-like signals. More importantly, Fig. 9 shows that also for the individual subsections there is again no dependence observable (within the statistical error of 10 mrad) between the measured phase shift at different time delays, which is the most important prerequisite for precision Ramsey-frequency comb spectroscopy.
6. Conclusion

The presented OPCPA system produces FC pulse pairs of more than 5 millijoule pulse energy and with a programmable inter-pulse delay of multiples of the cavity round-trip time of the master oscillators (currently 8 ns). We have investigated the phase coherence of these pulse pairs up to hundreds of nanoseconds, but extension to even tens of microseconds seems feasible. With the help of numerical simulations and an experimental investigation, it was established that despite absolute differential phase shifts of up to a few hundred milliradians (depending on the amplifier alignment), the differential phase shift remains constant within at least 10 mrad, as long as the pump pulse intensity ratio is stabilized to a few percent. In addition, within the statistical error of about 10 mrad, no dependence of the differential phase shifts on the inter-pulse delay could be observed. Given the statistical error as an upper bound for a potential phase shift, we estimate a maximum frequency error of 5 kHz in a Ramsey-type measurement (at the fundamental wavelength of the Ti:Sapphire FC). However, this upper bound was derived from a noise-limited measurement over a maximum inter-pulse delay of 336 ns. A phase measurement over longer pulse delays (such as microseconds) could potentially reduce this value proportionally to the delay time.

Furthermore, the new possibility to obtain Ramsey-like signals at programmable delays could also be used to measure multiple transitions at the same time. Similar to the well-known technique of Fourier transform spectroscopy [26, 27], complex excitation spectra can be retrieved from the combined Ramsey-signals via a Fourier-transform. Together with nonlinear upconversion of the amplified FC comb pulses, the combined Ramsey-signals will potentially enable multi-transition Ramsey-like spectroscopy in the extreme ultraviolet at the kHz-level.

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