The KMLabs RAEA for Carrier Envelope Phase (CEP) Stabilization Applications

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The KMLabs RAEA ultrafast laser amplifier system provides unparalleled high average power, performance and flexibility for an ultrafast ti:sapphire laser. As the next-generation, ruggedized successor to the KMLabs Dragon and Wyvern series lasers, we expect the RAEA to perform well for applications that requires CEP stabilization. Furthermore, the RAEA-HP-SP is now unique in the market as a high-power multipass ti:sapphire amplifier—an architecture that is far more extensively proven for CEP stabilization applications as compared with the more-common regenerative amplifier architecture (as is used in the RAEA HP). Here, we discuss the possible use of the RAEA for these applications. KMLabs will support the use of the RAEA laser for CEP stabilization applications, with the understanding that CEP stability is highly sensitive to the laboratory environment, making it unfeasible to offer guaranteed CEP stability specifications.

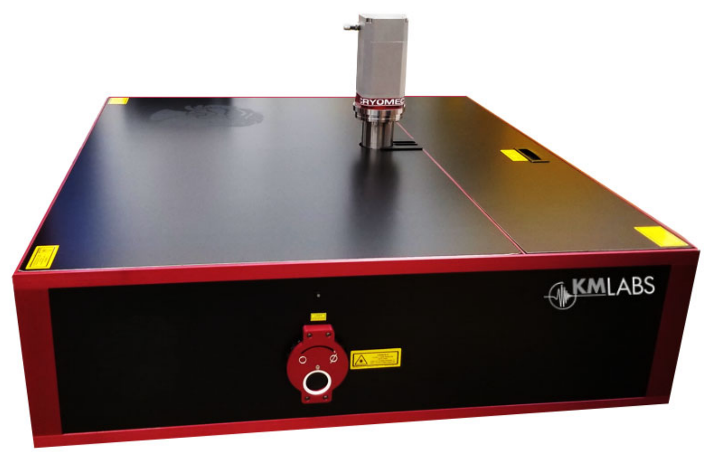

Figure 1: KMLabs is the leading supplier of custom high-performance ultrafast ti:sapphire lasers. KMLabs has developed a second-generation industrial-grade ultrafast ti:sapphire laser system, the KMLabs RAEA. Designed to provide the ultimate performance achieveable from a mJ-class ti:sapphire laser, its ultra-stable one-box construction, cryogenic cooling, and elegent design make it the ultimate laser source for high-harmonic applications.

1. Introduction

In modern day physics, many researchers study quantum systems with events occurring on the attosecond time scale. To measure these attosecond dynamics, physicists employ a variety of spectroscopic techniques using ultrafast pulsed laser sources. Analogous to stroboscopic photography for capturing slow-motion videos, the pulse length of the laser typically defines the fastest event that can be "resolved" by pump-probe spectroscopy techniques. However, for pulse durations of a few tens of fs or below, the pulse itself consists of just a few optical cycles. Under these conditions, the magnitude of the peak electric field varies quite significantly from pulse-to-pulse, as depicted in Figure 2(a). In this case, the cycles become distinguishable, and a variety of experimental approaches have been devised that make use of individual cycles to excite or

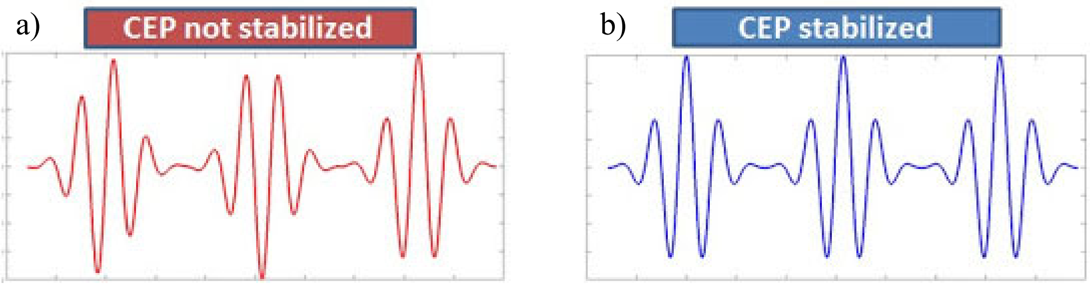

Figure 2. Time-dependent electric field of an ultrashort pulsed laser: (a) CEP **NOT** stabilized and (b) CEP stabilized.

interrogate a process. This strategy has made it possible for the first time to probe dynamics on sub-optical-cycle timescale, giving rise to the term *attosecond science.*

Much of attosecond science originated with the physics of high-order harmonic generation (HHG), where the field-driven dynamics results in generation of coherent light in the extreme ultraviolet (EUV) region of the spectrum. HHG is very sensitive to the cycle-by-cycle shape of the driving field, including to the phase of the "carrier," or optical field oscillations, with respect to the somewhat more slowly varying "envelope" of the laser pulse. In some experimental applications, if the phase within the carrier envelope can be stabilized from pulse-to-pulse, as drawn in Figure 2(b), then experimental interpretation can be made more straightforward. Conditions under which this relative phase is controlled from pulse-to-pulse is referred to as carrier-envelope phase (CEP) stabilized.

CEP stabilization can be shown to be equivalent to controlling individual mode frequencies of a modelocked laser, the so-called "frequency comb" that was recognized in the 2005 Nobel Prize. A frequency comb stabilized laser *is* a CEP-stable modelocked laser, and these lasers are now widely used in precision spectroscopy and optical clock technology. However, to access attosecond dynamics—which are inherently nonlinear and complex in nature, it is necessary to generate much higher energy pulses than are available from a frequency comb.

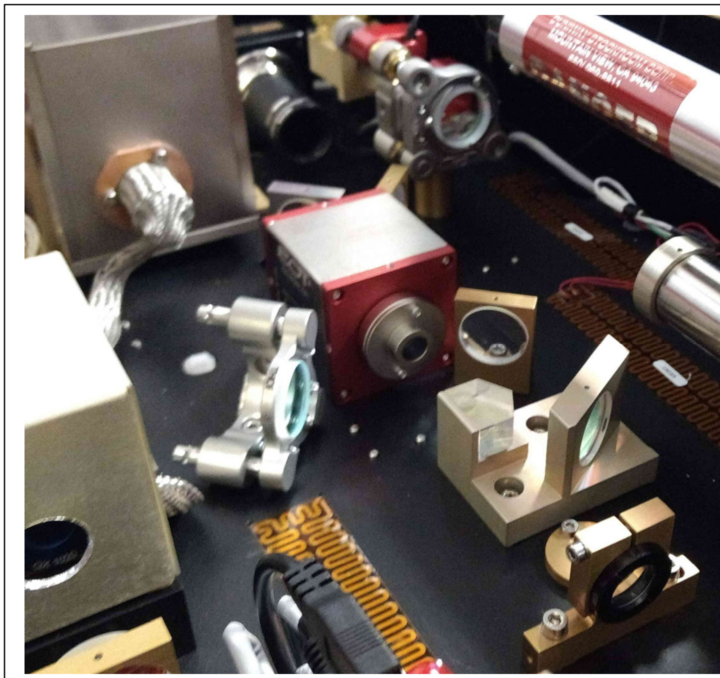

Figure 3: The RAEA ultrafast laser features a highly-engineered, modular construction with temperature control and thermal management. Many of the aspects of laser design that we have implemented for thermal load reasons also make the construction appropriate for CEP-stabilized operation.

Frequency comb stabilization technology is an area that has come a long way in terms of reliability, due to substantial R&D investments made in the past two decades. These lasers can be used as a seed source for an ultrafast laser amplifier system, resulting in high-intensity CEP-stable pulses for attosecond experiments. However, this amplification process is challenging to accomplish since the nature of the chirped-pulse amplification (CPA) process means that the CEP of a laser pulse injected into an amplifier is changed in a way that is exquisitely sensitive to changes in the amplifier optical path. *This requires that critical subsystems in the optical amplifier setup be interferometrically stable on short time-scales, and that additional feedback control be provided to adjust for thermally-induced (sub-micron) changes in the optical configuration.*

Nevertheless, despite the challenges, CEP stabilization of ultrafast amplifier systems can provide substantial benefits that are worth the effort to the researcher willing to push the limits. However, there are very few lasers on the market that can offer the features necessary to achieve CEP stabilization. Even with those features, control of the laboratory environment is daunting, but KMLabs has created an amplifier in the RAEA that is designed with CEP stabilization in-mind. The KMLabs RAEA amplifier—as pictured in Figure 1—is a one-box, high-power, cryogenically-cooled ultrafast ti:sapphire laser amplifier system. The two "flavors" currently offered are the RAEA HP regenerative amplifier, and the RAEA HP-SP multipass amplifier system. These two flavors are an update of our legacy Wyvern and Dragon amplifier systems, which have been discontinued. The RAEA offers a modular approach that changes only the amplifier module itself between these two flavors, retaining the same layout: oscillator, stretcher, compressor, and pump laser. The performance tradeoff between these two flavors is also simple: the RAEA HP is more efficient, offering more output average power, but with a longer pulse duration, ~35 fs as measured by rigorous FROG measurement. The RAEA HP-SP has a ~30% lower output power specification, but also a ~30% shorter pulse duration, ~25 fs. Other specifications are very comparable. These very short-duration pulses are often appropriate for high-harmonic generation applications and, consequently, for attosecond science research.

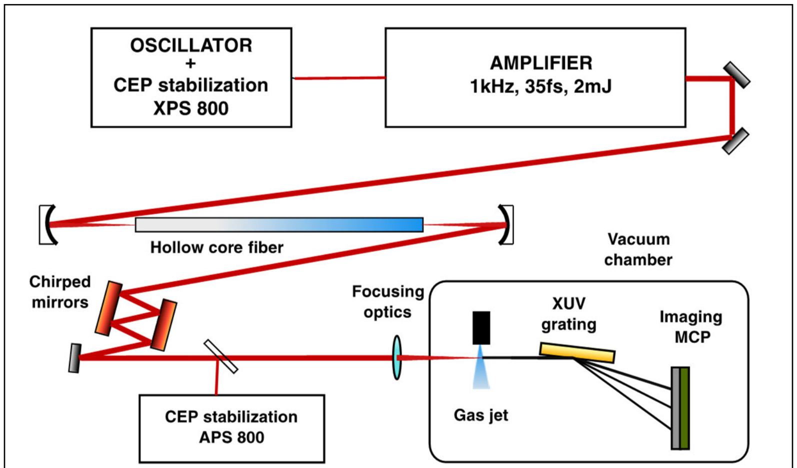

Figure 4: Configuration of CEP-stabilized Dragon laser amplifier system. Modifications to the system for CEP stability include: 1) modification of the Ti:sapphire oscillator for CEP stabilization using a MenloSystems XPS-800 system; isolation of the system from vibration that couples into the CEP of the system, and 3) monitoring the CEP of the output using an f-2f interferometer system; MenloSystems APS800 unit or similar. Figure from Phys Rev A 91(6), 8 (2015); dx.doi.org/10.1103/PhysRevA.91.063421

In comparison with the previous generation KMLabs Wyvern and Dragon systems, the RAEA is built using a rugged, modular configuration, including many changes in construction that directly apply favorably to its use for CEP stabilization. The modular design uses temperature controlled baseplates and an ultra-stiff design with a beam path very close to the baseplate (figure 3). This raises the frequency of

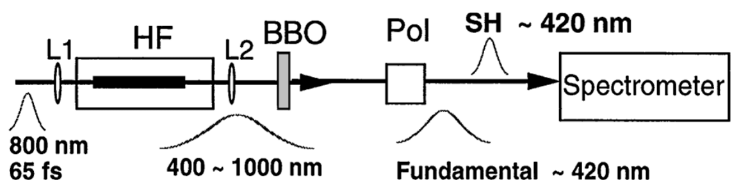

Figure 5: Configuration of spectral interferometry method for obtaining CEP of the output of a Ti:sapphire laser amplifier. L1, 250-mm focal length lens; L2, 600-mm focal length lens; HF, hollow fiber; BBO, 300-mm-thick BBO Crystal; Pol, polarizer. In the first step, the spectrum of the pulse is broadened—here this is done with a hollow fiber; however, white-light generation in a crystal such as YAG or sapphire works equally well. Frequency doubling of this beam then yields spectral interference fringes whose position reflects the CEP of the pulse. Monitoring the fringe position, can be used for a "slow" feedback to vary the oscillator CEP set point to keep the output at a stable value of output CEP.

vibrational resonances, reducing coupling of outside vibrations into the system.

2. Past results on CEP stabilization of KMLabs lasers

KMLabs has considerable successful experience with CEP stabilization of its Dragon laser platform, including several published works as well as supplemental data. This experience can be applied directly to the RAEA platform. The basic configuration of a CEP-stabilized KMLabs laser is well-illustrated in figure 4, taken from a publication by our customer[2] (this setup includes a hollow fiber pulse compressor, which is often used to further compress the pulses, but not required). Here, a KMLabs Griffin laser oscillator is modified by using a somewhat higher-power, low-noise pump laser, and by including mirror actuation that can be used to control the CEP of the oscillator output (a homebuilt version of the Griffin laser design was indeed used in the very first, Nobel-winning frequency comb experiments). The CEP is monitored using an f-2f interferometer (model XPS-800 by Menlo Systems), which provides the actuation signals to stabilize the CEP. These signals are injected into the amplifier, with a second stage of CEP monitoring provided by a simpler f-2f interferometer unit (model APS-800 by Menlo Systems), that provides a "slow" feedback to compensate for drifts in the CEP caused by thermal-expansion in the amplifier.

In our continuing efforts to deliver a "hands-off" amplifier system, the heart of the RAEA, the Ti:sapphire oscillator, was re-engineered to a closed-box solution called the Stryde, providing improved stability and reliability. The Stryde can be readily modified to include the elements necessary for CEP stabilization, and its small footprint allows for an f-2f interferometer (such as the Menlo Systems XPS-800) to be contained within the RAEA enclosure. The slow feedback—the APS 800 in figure 4 above—would be located external to the RAEA, ideally near the experiment. The Menlo systems APS 800 system is quite simple, and KMLabs has a design for this function that we are willing to share with customers (figure 5). A simple in-line setup can provide a fringe pattern indicative of the CEP of the laser output, that is subsequently used to set the target CEP value for the XPS-800. A variety of other techniques, such as Stereo ATI[3] and similar,[4] and the high harmonic process itself,[2] can be used to monitor CEP, even on a shot-by-shot basis at up to 100 kHz repetition rate.[5] KMLabs' recommendation is that, once the Ti:sapphire oscillator is stabilized, any long-term drift in the CEP of the output needs to be considered as integrated with the experimental setup itself. Building the f-2f system is an excellent experimental starting point for the customer to gain expertise. Furthermore, the very high repetition rates possible with the RAEA laser make it possible to further advance this technology, by providing faster feedback and possibly even providing primary CEP feedback for the entire system, circumventing the need for the XPS 800.

Figure 6 shows example CEP data from a KMLabs Dragon laser system, published in.[6] This publication also is a useful reference in that it identified the major sources of CEP fluctuation as vibrations entering the system, originating primarily from the pump laser and coupling into the stretcher and compressor. The vibration isolation used to obtain best performance is readily implemented in the RAEA platform: KMLabs can supply a RAEA with sorbothane mounts for the Stryde oscillator, stretcher, compressor, and pump laser, on request. Care must still need to be taken in maintaining a quiet, temperature stable environment, since acoustic noise is also a contributor to CEP noise. We would also note that the data of Figure 6 represent one of the more challenging implementations of CEP stability yet demonstrated.

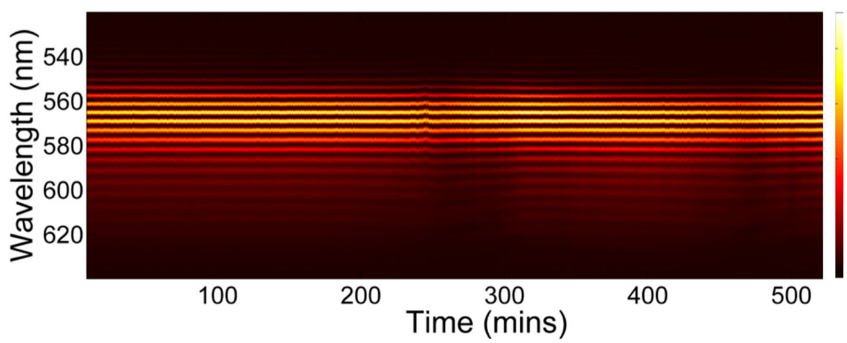

Figure 6: Raw data from f-2f interferometer from a KMLabs 2-stage, 1 kHz, 20 mJ (0.8 TW) laser system. The long-term stable fringe pattern illustrates a high degree of control over the CEP. In this laser, the CEP stability was measured to be <300 mrad.

In the lab, we have generally found that single-stage amplifiers are relatively quickly CEP stabilized, with 2-stage systems being much more challenging as interferometric stability is required over a much larger, nearly full-optical table setup. Figure 7 shows internal data from a single-stage KMLabs Dragon running at 1.2 mJ pulse energy and 3 kHz repetition rate. The RMS CEP stability of 100 mrad, measured on a single-shot basis, demonstrates the potential for excellent CEP performance from KMLabs lasers.

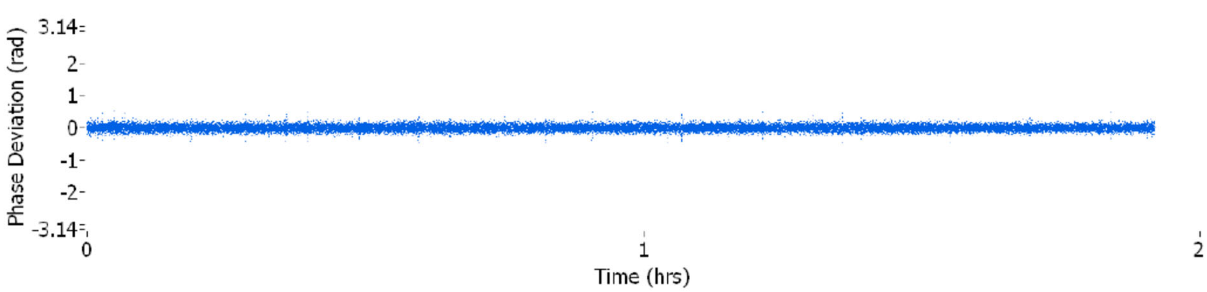

Figure 7: CEP stability from KMLabs Dragon laser, taken in 2008, running at 1.2 mJ output, 3 kHz, 27 fs. The shot-to-shot RMS fluctuation of 100 mrad from a single-stage amplifier system illustrates the potential for very high-performance CEP-stabilized systems.

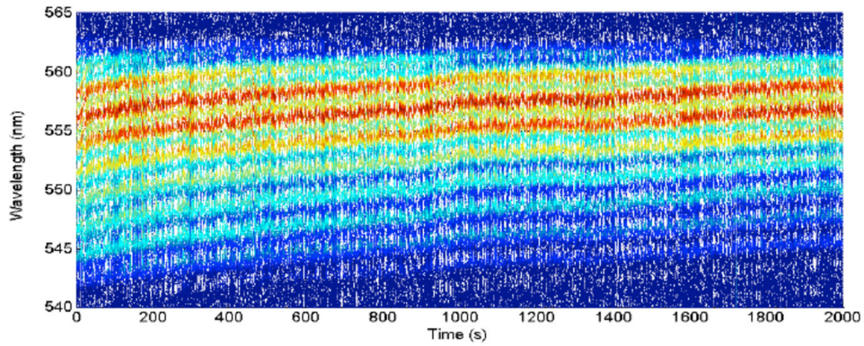

Figure 8: CEP stability from KMLabs Dragon laser, taken in 2008, running at 1.2 mJ output, 3 kHz, 27 fs, in the case where the oscillator CEP is stabilized but no feedback is applied from the amplifier f-2f interferometer. These data show that once the oscillator is stabilized, CEP drift from the amplified output is generally slow.

Figure 8 shows a sample data set from the system of figure 7, including CEP stabilization of the laser oscillator but without any direct feedback from the amplifier CEP. One can see that in general, once the laser oscillator is stabilized, CEP stability of the amplifier is obtained, with a slow drift resulting to thermal fluctuations and mechanical drift.

2.1 Other considerations and questions

KMLabs has more than a dozen years' experience with CEP-stabilized lasers, and the RAEA laser was designed in part with the eventuality of CEP stabilization in mind. However, the market for CEP-stabilized lasers has plateaued or shrunk in recent years. CEP stabilization in the past has been difficult enough that researchers have found simpler alternatives to obtain similar or even the same physical data. Many attosecond science experiments simply do not require CEP stabilization, but rather simply require that the *relative* optical phase of the pump and probe beams be maintained stably. An example of this is the RABBIT technique, which can be used to characterize the attosecond pulse train inherent to the HHG process. Indeed, if one is probing a process that is truly attoseconds in duration, there is no need to limit data acquisition to a single isolated attosecond pulse. Figure 9 shows an example using a KMLabs Dragon laser system, where an excited-state decay of ~200 attosecond lifetime, for a highly excited state within a solid, was measured directly—the very first measurements of an intrinsic, rather than laser-driven, attosecond process. The only laser requirement was interferometric stability of

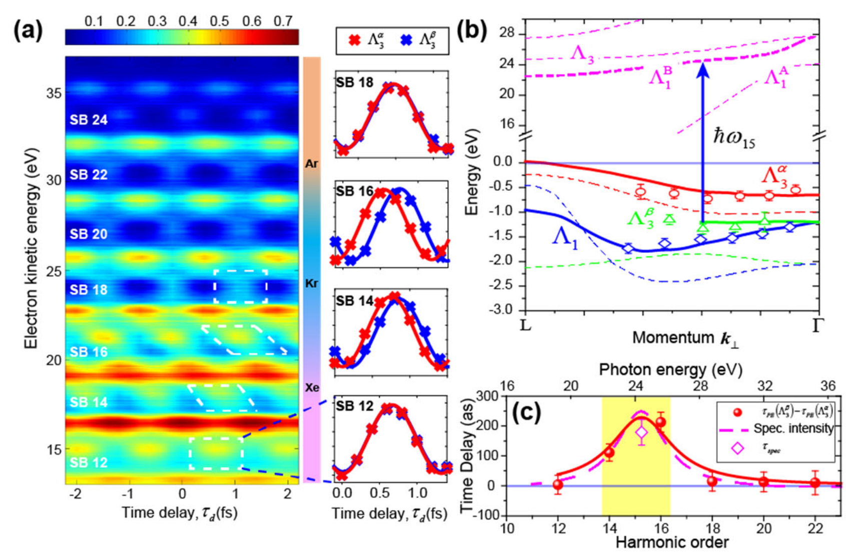

Figure 9: Data showing the first measurement of an attosecond time-scale excited-state lifetime—an intrinsic property of the material—using interferometric surface laser-assisted photoemission (RABBIT). From [1]

the pump and probe beams—not interferometric stability in the entire laser system, as is required for CEP stability.

Another approach that is vastly more tolerant of typical laboratory noise and environmental fluctuations is "CEP-tagging." In this approach, the CEP of each output pulses from a *standard*; i.e. not CEP-stabilized, laser is recorded on a shot-by-shot basis. Provided the experimental data are also acquired on a shot-by-shot basis (such as with COLTRIMS-type coincidence measurement experiments) and that the data acquisition system is synchronized, the CEP-dependent physics is randomly sampled, and can be sorted to reveal the CEP-dependent physics—with no great experimental variabilities and typically with higher CEP accuracy than is possible for CEP stabilization itself. Figure 10 illustrates the setup and data set obtained.

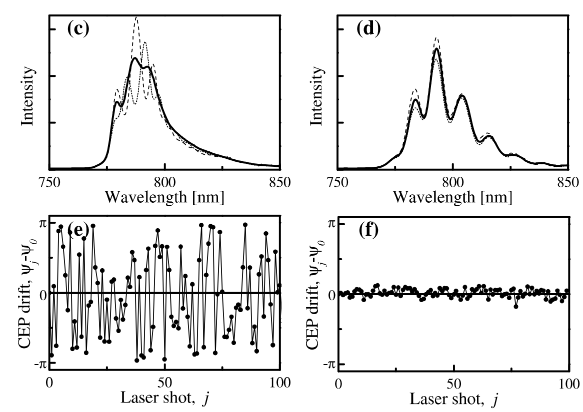

Figure 10: Spectral interference fringes, obtained using an apparatus such as in figure 5, from the signal (left) and idler (right) outputs, along with the CEP value obtained from the fringe variations. These data show that the signal wave exhibits random CEP, while the idler CEP is stable. From [7]

Finally, it would be noted that if *tunable* CEP-stable pulses are needed, which require the implementation of parametric frequency conversion, proper OPA configuration yields CEP-stable pulses *regardless* of the CEP stability of the driving laser. Specifically, the *idler* output from a white-light seeded OPA is inherently CEP stable, since in the difference frequency mixing process the CEP value cancels out (the OPA *signal* output carries all the CEP variation).[7] Thus, in the case of atomic or molecular photoionization experiments where tunable light is required in any case, and where the peak power requirement can be satisfied by the pulse energy and duration output of an OPA idler (or the second or third harmonic of this idler), CEP stability can be obtained passively. Figure 11 shows published results that illustrate the idler from an OPA is CEP stable.

Nevertheless, some experimental applications—particularly in chemical physics where the dynamics are in the few-femtosecond range (i.e. longer than the interval between pulses in an attosecond pulse train)—can benefit from CEP stabilization and single-attosecond pulse generation, and KMLabs is eager to support this work in commercially viable ways. We are also eager to work with pioneering customers to implement CEP stabilization on the RAEA platform, as it promises to provide fundamentally new capabilities, in particular for high repetition rate coincidence imaging. However, much of this ability is uncharted territory. Two issues to consider are:

a) Cryocooler implementation

The KMLabs Dragon CEP data as shown above all used complex vibration isolation schemes for the cryocooler, as competition drove a perception in the market that noise in the cryocooler would preclude CEP stabilization. Although these low-noise schemes proved compatible with CEP

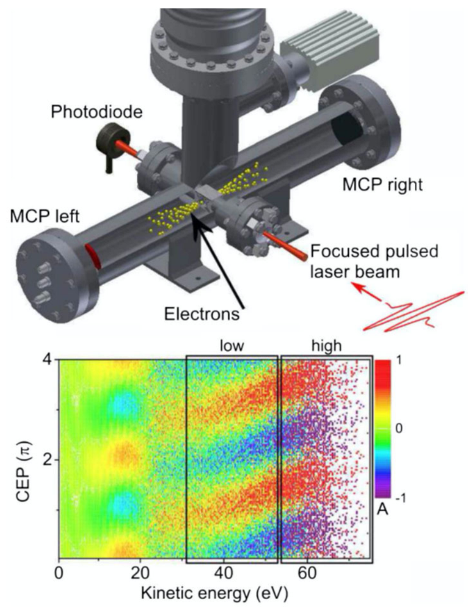

Figure 11: Schematic of the stereo-ATI apparatus for measuring CEP. Two TOF spectrometers measure the (left/right) electron spectra of Xe ionized by few-cycle laser pulses. The asymmetry (A) in the number of photoelectrons on the left (L) and right (R) detectors, A=(L – R)/(L+R) can be used to obtain the value of the CEP. From [5]

stabilization, they proved overly complex, significantly reducing the flexibility and reliability of the system.

In the work presented above, KMLabs established that in-fact the major sources of noise in the CEP stabilization *do not* seem to include the pulse-tube cryocooler used in the RAEA, but rather correspond to much higher frequencies. In follow-up, we did do simple tests for CEP stabilization using our most-standard cryogenic Ti:sapphire crystal mounting. The cryocooler can be switched-off for short periods while running the laser, allowing us to verify that the pulse-tube vibration adds negligible CEP noise to the output. *Thus, KMLabs recommends using the standard RAEA cryocell,* and anticipates no problems. Careful testing can verify the contribution of the (often nearly imperceptible) pulse-tube vibration on RMS CEP stability. In the event that vibrations from the cryocooler remains a significant contributor to CEP noise, the cryocell can readily be swapped for a vibration-free cold-helium cryocooler system with minimal disruption to the RAEA.

b) Regenerative amplifier vs multipass amplifier

The most significant uncertainty for CEP stabilization is in its implementation on the RAEA HP platform—the regenerative amplifier configuration—as opposed to the RAEA HP-SP multipass amplifier. All past KMLabs implementations of CEP stabilization have been done on the multipass amplifier. This configuration allows for amplification of a laser pulse by ~$10^{6\text{-}7}$ times with a total material path length of < 10 cm. To the extent that thermal variations change material properties and create drift in the CEP evolution of the pulse, the multipass amplifier is a superior platform for CEP stabilization.

In a regenerative amplifier, the material path length is much greater, since the beam makes multiple passes through a Pockels cell pulse switcher. Nevertheless, there are compelling reasons to implement CEP stabilization, specifically on *KMLabs'* regenerative amplifier systems. KMLabs is unique in offering Ti:sapphire regenerative amplifier systems that can run over a wide range of rep rates, including in the critical range of 10-200 kHz where the pulse energies can remain high enough to drive highly-nonlinear processes such as high harmonic generation, but where the higher repetition rates are amenable to precision studies.

Fortunately, there is reason to expect that CEP stabilization of the RAEA HP regenerative amplifier is possible and will provide unique capabilities for high repetition rate studies. In our work, for example as outlined in [6], the major sources of CEP noise are not in the amplifier itself, but in the pulse stretching and compression. Second, cases of CEP stabilized regenerative amplifiers are now found in the literature.[8] The caution here is that the vast majority of successful *experimental science* with CEP stabilized lasers has been done with multipass amplifier designs, from Femtolasers and from KMLabs, as is illustrated by the fact that[8] the original implementation of CEP stabilization on a Coherent laser amplifier has accumulated only 10 cites on ISI Web of Science® in 10 years. Nevertheless, the data in that paper demonstrate that there is no barrier in principle to CEP stabilizing a regenerative amplifier. Implementing this technique on a high repetition rate system—where the regenerative amplifier truly shines in performance—will provide capabilities not matched anywhere else on earth.

3. Summary and Conclusions

As a company with a heavy scientific emphasis and committed to expanding the capabilities of ultrafast lasers, KMLabs is eager to work with our enterprising customers on aspirations such as CEP stability. Nevertheless, the modest market for CEP-stable lasers, as well as the fact that the past major supplier of these lasers (Femtolasers GmbH, Vienna Austria) ultimately found it impossible to make a viable, profitable business in this area, necessitate that work in this area come with a customer commitment and a shared-risk approach. KMLabs believes that the RAEA can be the premier platform for CEP stable, high energy laser systems. To summarize KMLabs' interest in CEP stabilized systems:

- KMLabs is interested in working with leading research groups in demonstrating CEP stabilization of its RAEA laser system
- KMLabs has considerable experience implementing CEP stabilization in its Dragon laser systems. We have every reason to believe that the RAEA can provide equal or superior performance.
- KMLabs does not have experience CEP stabilizing its regenerative amplifiers. However, the literature indicates no fundamental problems.
- In the spirit of shared risk, KMLabs is willing to provide incremental modifications to its standard products to facilitate implementation of CEP stabilization *by the customer.* However, our warranty only extends to the "standard" (i.e. non-CEP stabilized) configuration of the laser.
- KMLabs is interested in committing to collaborative development, and we pledge to remain in close contact with the customer to support such a project. Telephone and e-mail consultations are free and not limited (with reasonable expectations in continuity of the dialogue; i.e. we will assign a primary point of contact to avoid repetition).
- On-site consultation and assistance on this aspect of the laser will be charged at our standard service rates; i.e. we are happy to help in this way, within our personnel scheduling constraints—but our preference is to implement this solution cost effectively in collaboration with a partner willing to gain the technical expertise necessary.

4. References

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