Final report **Grant No.** DE-FG02-09ER16115 DOE P **DOE Project No:** DE-SC0002378

Title: "PULSAR: A High-Repetition-Rate, High-Power, CE Phase-Locked Laser for the J.R. Macdonald Laboratory at Kansas State University"

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Synopsis: This instrumentation grant funded the development and installation of a state-of-the-art laser system to be used for the DOE funded research at the J.R. Macdonald Laboratory at Kansas State University. Specifically, we purchased a laser based on the KMLABs Red-Dragon design, which has a high repetition rate of 10-20 kHz crucial for multi-parameter coincidence measurements conducted in our lab. This laser system is carrier-envelope phase (CEP) locked and provides pulses as short as 21 fs directly from the amplifier (see details below). In addition, we have developed a pulse compression setup that provides sub 5 fs pulses and a CEP tagging capability that allows for long measurements of CEP dependent processes.

The PULSAR laser system:

Brief description: Main components. Two stage amplification. Continuous operation enabled by the use of a closed cycle He cooling system for the

CPA Characteristics:

Repetition rate: 10 kHz (or 20 kHz with half the energy/pulse)
Pulse Duration: < 21 fs amplifier output
Central frequency: 785 nm
Bandwidth: > 50-75 nm
Pulse energy: 2 mJ (at 10 kHz)
CEP stability: < 600 mrad for several hours

Pulse compression system: A hollow core fiber (HCF) and chirp mirrors compression system reduces the pulse duration from 21-23 fs down to 5 fs (and below). A beam pointing stabilization unit (model) is used to improve the coupling and input stability into the HCF. The resulting pulse characteristics are:

Pulse Duration: < 5 fs
Central frequency: ~740 nm
Pulse energy: 800 μJ
Bandwidth: > 200 nm

Laser beam diagnostics: Pulse characterization is accomplished using a SHG-FROG (Mesaphotonics FROGScan Ultra) at the CPA output and an Autocorrelator (Model GSAKF020k FEMTOMETER from FEMTOLASER) after the HCF pulse compression, a spectrometer (Ocean Optics USB2000+VIS-NIR-ES) and power meters (Coherent Fieldmate). The CEP is monitored by an f-2f interferometer and a stereo ATI CEP meter (used also for CEP tagging).

Beam Transport: The laser beam is transported to a few end stations where experiments focused on attosecond physics or control of simple atoms, molecules and surfaces are conducted. Given the high beam energy, the transport lines can be evacuated and use large diameter (2") optics.

CEP tagging: We have been involved, as part of a large international team, in the development of CEP tagging and in demonstrating its advantages over CEP locking for long timescale measurements. The proof of principle experiments were conducted at MPQ, Garching, Germany, taking advantage of their sub 5 fs laser pulses (see, e.g., Refs. [1-2]). Since then we have improved our pulse compression system and can produce similar short pulses at JRML. Moreover, our ATI stereo CEP meter is operational and has already produced publishable results (see, e.g., Pub. [1]).

Research employing PULSAR: Since its installation in August 2012, PULSAR has been used for research in parallel to the continuous development of some of its capabilities, such as transport lines, beam diagnostics, pulse compression, and CEP locking and tagging. Even though the main focus of this report is on instrumentation development, installation and specifications, it is useful to mention a few studies already conducted with this laser system, since that best illustrates how it has extended the research capabilities of JRML.

As a state-of-the-art CEP-locked laser system, PULSAR provides an excellent platform for technology advancement. For example, we have characterized the performance of an f-2f interferometer using a high-speed CMOS camera in single-shot mode with ~5-fs pulses from PULSAR. By using the stereo ATI CEP meter and the f-2f interferometer to simultaneously measure the CEP, we have determined that CEP-tagging can be performed with the much cheaper and easier-to-implement single-shot f-2f method (see Pub. [2]).

In terms of physics, one nice example of research made possible by this system is the measurement of the CEP dependence of dissociation of an H_2^+ beam. CEP dependence measurements of such extremely low density molecular-ion-beam targets require very long data accumulation times (8-24 hours at 10 kHz), and therefore preliminary attempts using our 1-2 kHz laser system with CEP locking failed. In contrast, we were able to successfully make the measurement by employing PULSAR. The importance of using the molecular ion as a target stems from our ability to theoretically treat this system in a strong-field laser nearly exactly, thus enabling us to make direct, unambiguous, quantitative comparisons between theory and experiment, and by so doing confirm our understanding of strong-field coherent control via the carrier-envelope phase (see Pub. [1] and Ref. [3]).

Another recent example of studies made possible by the new PULSAR laser system is a measurement of the CEP dependence of D* formation in D_2 fragmentation induced by 5 fs laser pulses. This project has resulted in three manuscripts, presently in preparation, and a few invited talks in conferences (Pres. [1,5]) and other institutions (Pres. [2-4]).

A few other projects using PULSAR beams are nearing completion (see Pubs. [2,3]), and some have already been presented in conferences (see Pres. [6-19]) and will be prepared for publication in the near future. These publications will all appear in the JRML research report on DOE grant No. DE-FG02-86ER13491, which supports our laser-matter interaction research.

References:

1. "Single-shot carrier-envelope-phase-tagged ion-momentum imaging of nonsequential double ionization of argon in intense 4-fs laser fields", Nora G. Johnson, O. Herrwerth, A. Wirth, S. De, I. Ben-Itzhak, M. Lezius, B. Bergues, M.F. Kling, A. Senftleben, C.D. Schröter, R. Moshammer, J. Ullrich, K.J. Betsch, R.R. Jones, A.M. Sayler, T. Rathje, K. Rühle, W. Müller, and G.G. Paulus, Phys. Rev. A **83**, 013412 (2011).
2. "Attosecond tracing of correlated electron-emission in nonsequential double ionization", Boris Bergues, Matthias Kübel, Nora G. Johnson, Bettina Fischer, Nicolas Camus, Kelsie J. Betsch, Oliver Herrwerth, Arne Senftleben, A. Max Sayler, Tim Rathje, Itzik Ben-Itzhak, Robert R. Jones, Gerhard G. Paulus, Ferenc Krausz, Robert Moshammer, Joachim Ullrich, and Matthias F. Kling, Nature Communications **3**, 813 (2012); doi: 10.1038/ncomms1807.
3. "Coherent control at its most fundamental: Carrier-envelope-phase-dependent electron localization in photodissociation of a H_2^+ molecular ion beam target", T. Rathje, A.M. Sayler, S. Zeng, P. Wustelt, H. Figger, B.D. Esry, and G.G. Paulus, Phys. Rev. Lett. **111**, 093002 (2013).

Publications:

1. "Carrier-envelope phase control over pathway interference in strong-field dissociation of H_2^+", Nora G. Kling, K.J. Betsch, M. Zohrabi, S. Zeng, F. Anis, U. Ablikim, Bethany Jochim, Z. Wang, M. Kübel, M.F. Kling, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak, Phys. Rev. Lett. **111**, 163004 (2013).
2. "Shot-to-shot correlation of the carrier-envelope phase measured by an f-2f interferometer and a stereo-above-threshold-ionization phase-meter", Xiaoming Ren, Aram Vajdi, Varun Makhija, Carlos Trallero and Vinod Kumarappan, Optics Express – in preparation.
3. "Rudimentary coherent control of H_2^+ dissociation by broad bandwidth laser pulses", M. Zohrabi, L. Graham, U. Lev, U. Ablikim, B.D. Bruner, J.J. Hua, J. McKenna, K.J. Betsch, B. Jochim, A.M. Summers, B. Berry, D. Strasser, O. Heber, Y. Silberberg, D. Zajfman, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak, Phys. Rev. A – in preparation.

Dissertations:

1. "Controlling the dynamics of electrons and nuclei in ultrafast strong laser fields", Nora G. Kling, PhD Dissertation, Kansas State University (2012).
2. "Quantum control of molecular fragmentation in strong laser fields", Mohammad Zohrabi, PhD Dissertation, Kansas State University (2014) – in preparation.

Presentations – invited:

1. "Carrier-envelope phase (CEP) control over molecular dynamics - two cases:"
   a) CEP control over pathway interference in strong-field dissociation of H_2^+,
      Nora G. Kling, K.J. Betsch, M. Zohrabi, S. Zeng, F. Anis, U. Ablikim, Bethany Jochim, Z. Wang, M. Kübel, M.F. Kling, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak,
   b) CEP control over D* formation in strong-field dissociation of D_2,
      M. Zohrabi, B. Berry, Nora G. Kling, U. Ablikim, Travis Severt, Bethany Jochim, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak,
   Itzik Ben-Itzhak
   Department of Energy, Atomic, Molecular and Optical Sciences Research meeting,
   Bolger Conference Center, Potomac, Maryland, October 2013.
2. "Carrier-envelope phase control over molecular dynamics in strong-field laser pulses",
   Itzik Ben-Itzhak, Nora G. Kling, M. Zohrabi, B. Berry, U. Ablikim, B. Jochim, T. Severt, K.J. Betsch, K.D. Carnes, Shuo Zeng, F. Anis, J. Hernández, Yujun Wang, and B.D. Esry,
   Colloquium,
   Department of Particle Physics and Astrophysics, Weizmann Institute of Science, Rehovot, Israel, January 2014.
3. "Carrier-envelope phase control over molecular dynamics in strong-field laser pulses",
   Itzik Ben-Itzhak, Nora G. Kling, M. Zohrabi, B. Berry, U. Ablikim, B. Jochim, T. Severt, K.J. Betsch, K.D. Carnes, Shuo Zeng, F. Anis, J. Hernández, Yujun Wang, and B.D. Esry,
   Physical Chemistry Seminar,
   Department of Chemistry, Tel-Aviv University, Tel-Aviv, Israel, March 2014
4. "Carrier-envelope phase control over molecular dynamics in strong-field laser pulses",
   Itzik Ben-Itzhak, M. Zohrabi, N.G. Johnson, B. Berry, U. Ablikim, B. Jochim, T. Severt, K. J. Betsch, K.D. Carnes, Shuo Zeng, F. Anis, J. Hernández, Yujun Wang, and B.D. Esry,
   Physical Chemistry Seminar,
   Department of Chemistry, Hebrew University, Jerusalem, Israel, May 2014

5. "Carrier-envelope phase control over molecular dynamics in strong-field laser pulses", Itzik Ben-Itzhak, M. Zohrabi, Nora G. Johnson, B. Berry, U. Ablikim, B. Jochim, T. Severt, K.J. Betsch, K.D. Carnes, Shuo Zeng, F. Anis, J. Hernández, Yujun Wang, and B.D. Esry, Gordon Research Conference (GRC) - Multiphoton Processes, Bentley University, Waltham, Massachusetts, USA, June 2014

Presentations – contributed[1]:

Annual Meeting Division of Atomic, Molecular and Optical Physics (DAMOP), Quebec City, Canada, June 2013

6. "Capabilities of the PULSAR ultrafast laser system for attosecond research", K.J. Betsch, Z. Wang, Nora G. Kling, M. Kübel, B. Langdon, D. Ramondson, M. Kirchner, C.W. Fehrenbach, K.D. Carnes, V. Kumarappan, C. Trallero-Herrero, M.F. Kling, and I. Ben-Itzhak.
7. "Exploring trends in strong-field dissociation of small polyatomic molecules", Bethany Jochim, M. Zohrabi, U. Ablikim, B. Berry, T. Severt, K.D. Carnes, I. Ben-Itzhak.

Ultrafast atomic and molecular physics with cutting-edge light sources: New opportunities and challenges, ITAMP workshop at Kansas State University, Manhattan, Kansas, USA, November 2013

8. "Carrier-envelope phase control over pathway interference in strong-field dissociation of H_2^+", Nora G. Kling, K.J. Betsch, M. Zohrabi, S. Zeng, F. Anis, U. Ablikim, Bethany Jochim, Z. Wang, T. Severt, B. Berry, M. Kübel, M.F. Kling, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak.
9. "CEP control over D* formation in strong-field dissociation of D_2", M. Zohrabi, B. Berry, Nora G. Kling, U. Ablikim, Travis Severt, Bethany Jochim, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak.

Annual Meeting Division of Atomic, Molecular and Optical Physics (DAMOP), Madison, Wisconsin, June 2014

10. "Combining phase-tagging and CEP-stabilization for increased precision in CEP-dependent measurements", Kevin D. Carnes, K.J. Betsch, A.M. Summers, I. Ben-Itzhak, B. Langdon, D. Raymondson, and M. Kirchner.
11. "Carrier-envelope phase dependences of D_2 dissociation into Rydberg deuterium fragments", M. Zohrabi, Ben Berry, U. Ablikim, Nora G. Kling, Travis Severt, Bethany Jochim, Shuo Zeng, D. Ursrey, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak.
12. "Three-body fragmentation of triatomic molecular ions in a strong laser field", U. Ablikim, M. Zohrabi, Bethany Jochim, Ben Berry, K.D. Carnes, and I. Ben-Itzhak.
13. "Carrier-envelope phase effects in state-selective measurements of D* fragments from D_2 molecules", Ben Berry, M. Zohrabi, U. Ablikim, Nora G. Kling, Travis Severt, Bethany Jochim, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak.
14. "Carrier-envelope phase control over pathway interference in strong-field dissociation of H_2^+ molecular ions", M. Zohrabi, Nora G. Kling, K.J. Betsch, S. Zeng, F. Anis, U. Ablikim, Bethany Jochim, Z. Wang, M.F. Kling, K.D. Carnes, B.D. Esry, I. Ben-Itzhak, and M. Kübel.
15. "Significant carrier-envelope phase effects for intense, many-cycle laser pulses", Yujun Wang, J.V. Hernández, M. Zohrabi, Ben Berry, U. Ablikim, Nora G. Kling, Travis Severt, Bethany Jochim, K.D. Carnes, I. Ben-Itzhak, and B.D. Esry.

[1] Speaker of contributed talk is underlined. In contrast, the presenter of a Poster is not underlined.

16. "Defining photon channels in strong-field physics: the photon-phase Fourier representation", Shuo Zeng, Mohammad Zohrabi, Ben Berry, Utuq Ablikim, Nora Kling, Travis Severt, Bethany Jochim, Kevin Carnes, Itzik Ben-Itzhak, and Brett Esry.
17. "Achieving a quantitative understanding of carrier-envelope phase effects in the benchmark H_2^+ molecule", Shuo Zeng, Nora Kling, Kelsie Betsch, Mohammad Zohrabi, Fatima Anis, Utuq Ablikim, Bethany Jochim, Zhenhua Wang, Matthias Kübel, Matthias Kling, Kevin Carnes, Brett Esry, and I. Ben-Itzhak.
18. "Strong-field dissociation of CS^{2+} via a pump/dump-like mechanism", T. Severt, M. Zohrabi, K.J. Betsch, U. Ablikim, Bethany Jochim, K.D. Carnes, S. Zeng, B.D. Esry, I. Ben-Itzhak, and T. Uhlíková.

Gordon Research Conference (GRC) – Multiphoton Processes, Bentley University, Waltham, Massachusetts, USA, June 2014

19. "Laser induced charge asymmetric dissociation (CAD) in the transient CD dication", L. Graham, M. Zohrabi, B. Gaire, U. Ablikim, B. Jochim, B. Berry, T. Severt, K.J. Betsch, A.M. Summers, U. Lev, O. Heber, D. Zajfman, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak.
20. "Carrier-envelope phase control over D* formation in strong-field dissociation of D_2", M. Zohrabi, B. Berry, U. Ablikim, Nora G. Kling, Travis Severt, Bethany Jochim, K.D. Carnes, Shuo Zeng , D. Ursrey, J. V. Hernández, Y. Wang, B.D. Esry, and I. Ben-Itzhak.