Final project Report

Spatio-temporally shaped deep UV laser source for ultrabright photocathodes using novel upconversion techniques

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Sponsoring Program Office: USDOE Office of Science

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Major goals and objectives of this project

The goal of this project was to take a VUV laser system designed by Kapteyn-Murnane Labs (KMLabs)—the Hyperion VUV, and increase its functionality, efficiency, and applicability by further probing the fundamental physics taking place in the system and optimizing the system performance. More specifically, DOE accelerator applications—particularly for XFELs that require low-emittance electron beams, require precisely controlled ultrashort pulses in the deep-UV at 250-260 nm to drive the photocathode electron source. This is conventionally done using an ultrafast infrared laser, with subsequent stages of nonlinear upconversion in solid-state nonlinear-optical crystal media (principally BBO, which is commonly used for UV upconversion). However, maintaining high spatial mode beam quality, as well as precisely controlled temporal profile, has proven a persistent challenge.

As a possible alternative to this approach, KMLabs has demonstrated a system that generates vacuum ultraviolet light (VUV) by upconverting pulses from an infrared ultrafast laser in a *gas-filled* hollow waveguide, driving a process of highly cascaded harmonic generation (HCHG). Because HCHG upconversion is done in a gas, rather than a solid, at high intensity in a guided-wave geometry, it retains an excellent beam spatial profile as well as a large spectral bandwidth that allows for control over the temperal structure of the pulse. A challenge, however, is in energy scaling of this HCHG technique to the ~1-20 µJ pulses required for photocathode applications. To-date, the pulse energies used in published HCHG work are sub-10-nJ.[1] This project proposed to explore ways to generate higher-energy pulses using HCHG, as well as to investigate spectral and temporal shaping of the deep UV pulses by pulse shaping the infrared laser used to seed the process.

When the proposal was initially written, the intent was to use the KMLabs-built Y-Fi™ ultrafast fiber laser to drive the process. However, this product line was acquired by Thorlabs, Inc. Thorlabs has established a laser division in Colorado, bringing additional photonics commercial activity to the state. However, this acquisition also meant that KMLabs no longer has a development program for this laser, and no ability to customize the laser for the HCHG application. Specifically, this development left us with no readily accessible route for investigating pulse shaping for spectral control of HCHG output. Instead, KMLabs decided to work with the Kapteyn-Murnane research group at JILA, who had acquired a Light Conversion Pharos laser (20W output, up to 1 MHz repetition rate, ~200 fs pulses) suitable for driving the HCHG process. This resulted in a delay and no-cost extension, as well as redirecting the project to study energy scaling and conversion efficiency of the process specifically to the 259 nm harmonic.

In this project during the time of the NCE, the experimental work primarily focused on goal #3, which was to measure power and conversion efficiency, and to optimize the process for maximal conversion efficiency and pulse energy to 259 nm. In particular, the ability of the Pharos laser to generate higher initial IR pulse energies (albeit at lower pulse repetition-rate) made it attractive to try to scale the HCHG process to higher energies. The milestones listed in the original funded proposal are summarized below:

MILESTONE	Phase and Personnel	Timeline (mo)
Bring prototype Y-Fi VUV HCHG laser back into operation	Phase I, KMLabs	1-3
Design of spectral pulse shaper for 1040 nm	Phase I, KMLabs	1-2
Power and M^2 measurements of 350 nm and 260 nm output	Phase I, KMLabs	2-5
Incorporate spectral shaper into laser system	Phase I, KMLabs	6
Validate fidelity of spectral shaping of deep UV light using shaper	Phase I, KMLabs	6-9
Implement 350 nm third harmonic generation of fiber laser output using upconversion in crystals	Phase I, KMLabs	9-12

The approach we used for energy scaling of the HCHG process to generate light at 259 nm was to compare the cascaded process with single-step four-wave mixing upconversion. The difference is as follows:

1. In HCHG, two wavelengths of light, 1034 nm and its second harmonic (generated conventionally using BBO second harmonic generation) are injected into a hollow-core antiresonant photonic crystal fiber (PCF). Nonlinear mixing of these two waves then produced 345 nm through a phase matched four-wave difference frequency mixing process ($2\bullet2\omega - \omega = 3\omega$), subsequently followed by a second four-wave mixing step to generate 259 nm through ($2\omega + 3\omega - \omega = 4\omega$).
2. Alternatively, the conventional third-harmonic upconversion using BBO can be done with upconversion efficiency from 1034 nm to 345 nm of 15-25%. By injecting a higher energy 345 nm pulse into the waveguide, along with 517 nm, the process ($2\bullet3\omega - 2\omega = 4\omega$) can generate 259 nm.

Our hope was that by employing only the last step of conversion in the waveguide, we could increase the pulse energy at 259 nm to values relevant to photocathode development. This approach seemed very attractive: third-harmonic generation to 345 nm using BBO crystals is still in the regime where sum-frequency upconversion is very efficient, due to modest values of group-velocity walkoff. In the lab, we could readily obtain several watts average power at 1 MHz at 345 nm. In comparison, the first step of the HCHG process results in beam power of ~ 300 mW. If using an increased 3ω power could translate directly into an increased pulse energy from the PCF upconversion step, this would be a significant advance.

b. What was accomplished under these goals?

During this project, we developed an entirely new setup for studying HCHG in hollow photonic crystals, starting with a Light Conversion Pharos laser to generate pulses at 1034 nm, 190 fs, and up to 20W output power. This beam was then split into three lines—the first employing the 1034 nm directly, the second frequency doubled to 517 nm, and the third frequency-tripled to 345 nm. For each of these steps, we worked hard to maintain a near-Gaussian spatial mode for the beam. The lines included appropriate variable telescopes to control the beam size, as well as adjustable delays. In this way, each color—1034 nm, 517 nm, 345 nm—could be optimized to couple efficiently into the PCF waveguide with timing overlap. This setup was operated at a 1 MHz repetition-rate. The Pharos laser can still provide 20W output at 100 kHz, with an order-of-magnitude higher

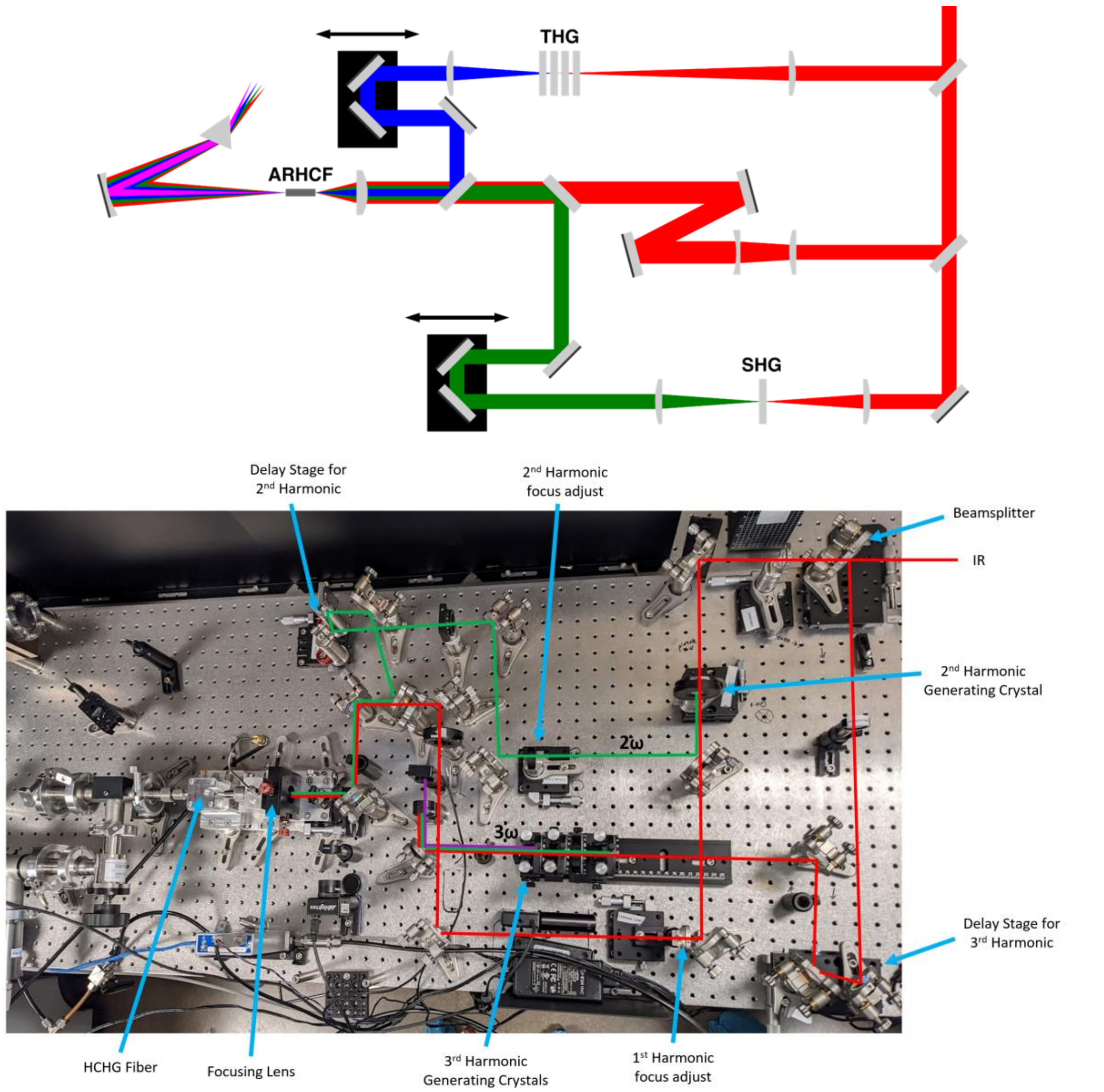

Figure 1: Optical layout for multi-wave mixing in a gas-filled hollow photonic crystal waveguide, and photo of setup. This setup was designed to explore the scaling fundamentals of the wavemixing process, with the SBIR-relevant objective to optimize pulse energy at 259 nm.

pulse energy. This provided us with excess pulse energy to explore parameter space—the setup also includes waveplate/polarizers to individually adjust pulse energy to explore parameter space. Figure 1 shows a photo of the apparatus.

Optimizing frequency conversion with excellent mode quality was straightforward for the 2ω beamline, but the 3ω beamline required considerable effort. Nevertheless, conversion efficiencies exceeding 15% with TEM_{00} mode quality were obtained—in the current configuration, 3ω pulse energies of nearly 1.1 μJ were generated. The table of figure 2 shows the resulting conversion efficiencies.

	Input (W)	**Output (W)**	**Efficiency**
2ω	8.70	3.77	43%
3ω	6.13	1.07	17.5%

Figure 2: Frequency conversion of LC Pharos laser to near-TEM_{00} spatial mode. Rep-rate 1 MHz.

The next step was to optimally couple each of these beams into a 25 μm-core hollow PCF waveguide. This process takes considerable practice and expertise, undertaken by new PhD student Jeremy Thurston assisted by Postdoc Dr. Tika Kafle and by the KMLabs team (primarily, Liam Weiner). He developed a systematic optimization procedure for coupling each color into the waveguide separately, and then revised this procedure to ensure the same focal position for each beam, allowing for simultaneous optimization of all three colors into the waveguide. This setup makes possible an “apples to apples” comparison of upconversion efficiency for the two-stage HCHG vs single-stage four-wave-mixing process—and even to try three-color wavemixing. Figure 3 shows the excellent results of optimal coupling of ω, 2ω, and 3ω into the PCF. Coupling efficiency exceeding 90% was possible for the second- and third-harmonic light, with a high-quality exit mode.

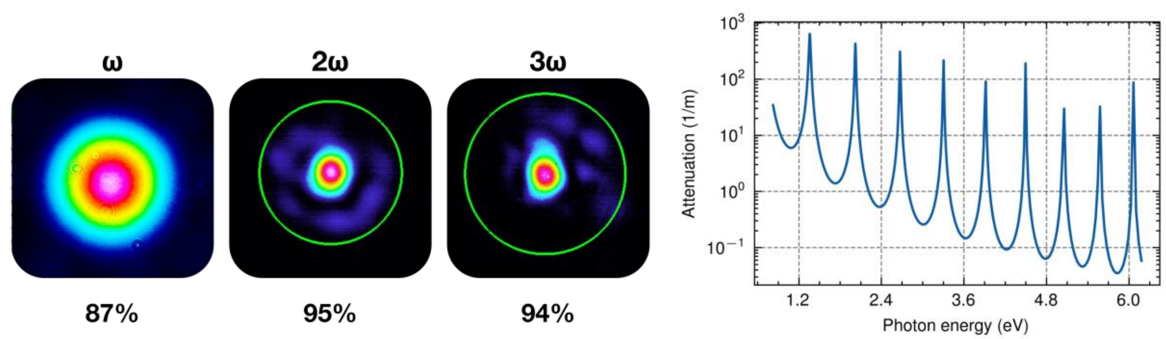

Figure 3: Results from coupling of 1034 nm, 517 nm, and 345 nm light into a 6 cm length of 25 μm mode-field diameter antiresonant hollow PCF fiber. Excellent coupling efficiency for each of these colors ensures an optimum nonlinear interaction between the colors. This outcome is consistent with calculation of loss in this antiresonant hollow-core waveguide[doi.org/10.1364/JOSAB.36.000069], which shows that the 2w and 3w waves (at 2.4 eV and 3.6 eV) see low attenuation, with slightly higher loss at the 1.2 eV fundamental.

Following this coupling optimization, the PCF was then put into a gas cell backfilled with Xenon, which is the optimal nonlinear-optical gas medium for HCHG. In studies of 4w conversion efficiency, the light generated could exit through a window, and was separated spectrally from the pump light using a prism. Pressure tuning could then be used to optimize the nonlinear process. Figure 4 shows pressure-dependent tuning of the 4ω output power. Figure 5 shows the mode emerging from the waveguide. The spectrum of the pulse is a 50 meV FWHM—limited by the resolution of the spectrometer. The time-bandwidth limit for this spectrum corresponds to ~40 fs, which is somewhat shorter than might be expected for ~200 fs driving laser pulses. For high spectral resolution scans in the VUV spectral region, a Czerney-Turner monochromator designed and built by KMLabs was installed. This configuration is designed to record either high throughput or high-resolution spectra from 120~250 nm. Figure 6 is a spectrum recorded with this monochromator in the high throughput configuration with an estimated spectral resolution on the order of 100 meV.

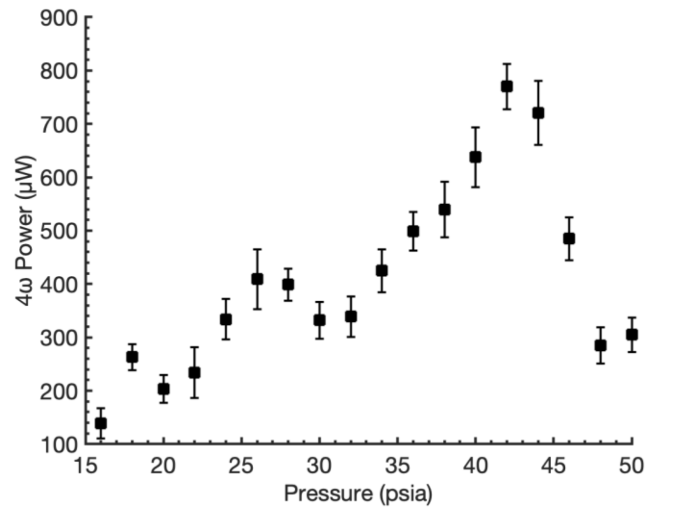

Figure 4: Pressure tuning for 4w generation in PCF, with 3 W input of light at 517 nm, and 1 W at 345 nm. The curve shows a clear phase-matched optimization at relatively high pressure in Kr (45 psia~2300 torr).

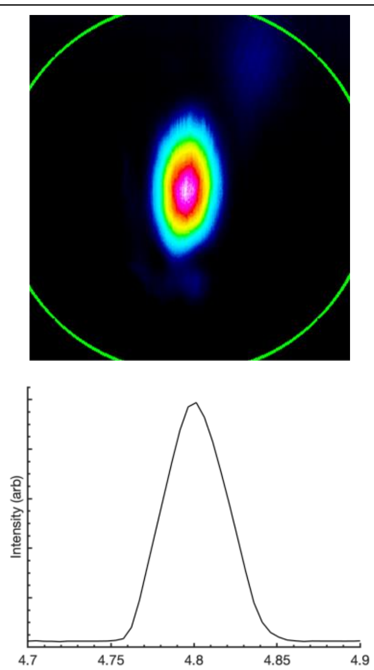

Figure 5: Spatial and spectral mode of 4w light generated in the PCF hollow waveguide.

The data of figure 4 were optimized for the highest pulse energy into the PCF waveguide for the 2w and 3w light (3 μJ and 1 μJ, respectively) that could be coupled into the waveguide while allowing for long-term operation and characterization; i.e. higher pulse energies resulted in rapid damage to the waveguide. The power output, 0.8mW = 0.8 nJ per pulse at 1 MHz, however, remains only comparable to the pulse energy obtained through cascaded FWM with ω, 2ω coupling into the PCF (6 nJ pulse energy). Thus, although we showed that this approach works for generating light at 259 nm, the total pulse energy achieved seems limited to approximately the same value.

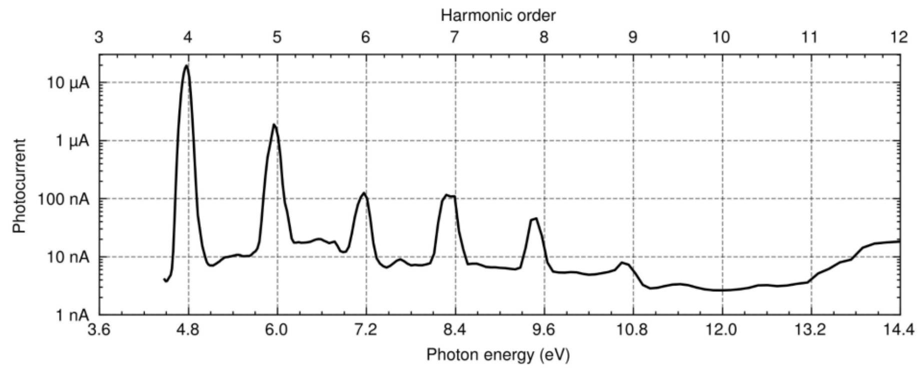

Figure 6: HCHG spectrum recorded in Czerney-Turner monochromator. Inputs are 1.5 uJ of 1030 nm and 1.5 uJ of 515 nm at 1 MHz and 90 fs. Overall throughput drops around 10 eV, partially due to the drop in efficiency of Al/MgF2 optics at these wavelengths.

To attempt to understand these results, we used the PyNLO code [2, 3] to simulate the upconversion process in both cases. The results are shown in figure 7. These simulations are broadly consistent with our experimental results. The top graph shows a very comparable output pulse energy at 4ω (4.8 eV) when injecting 2ω and 3ω into the waveguide, vs injecting ω and 2ω. The bottom graph shows experimental results using all three colors injected into the waveguide. This calculation shows that multicolor driving

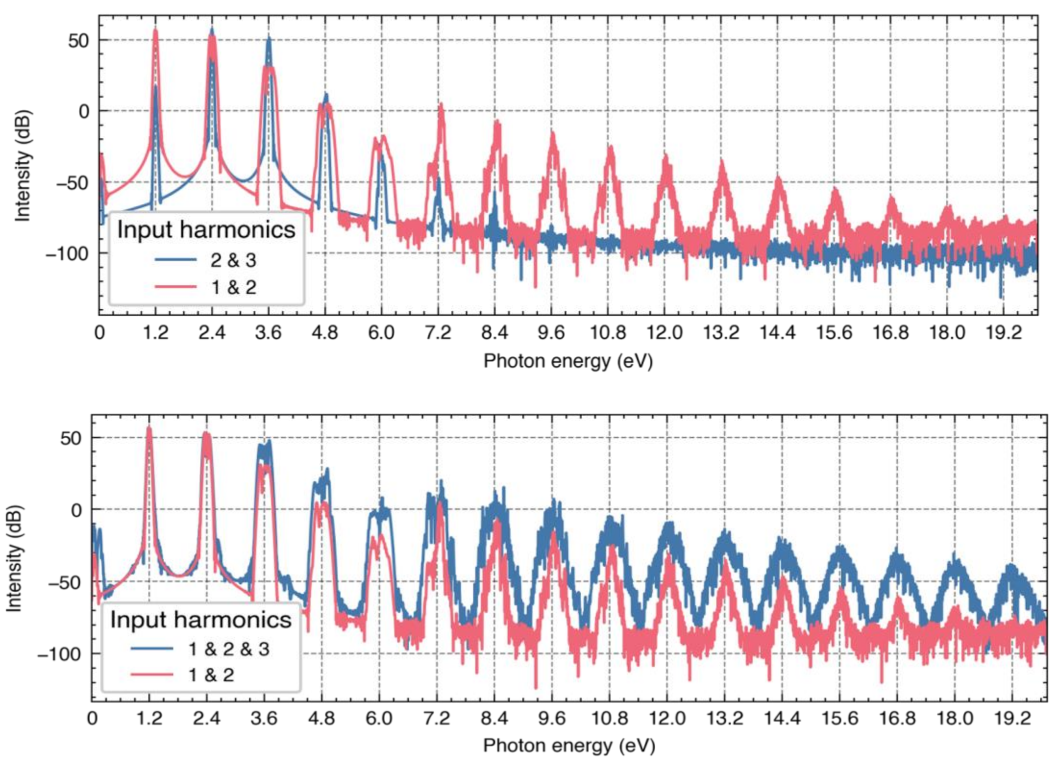

Figure 7: PyNLO simulations of multi-wave mixing in PCF waveguides. Simulation parameters are: 25 µm spot size, 5 µJ @1034 nm, 3 µJ @ 517 nm, and 1 µJ @345 nm, with a 6-cm long fiber. These result show that the low-loss propagation of the 1034 nm light in the waveguide is critical for driving a strong cascaded upconversion process.

may well have benefits for extending this source to much shorter wavelengths—but that the increase is generally *not* significant for the generation of deep-UV light.

Future work

Although the SBIR project funds have been expended without reaching a goal sufficient to apply for phase II funding, further related work is proceeding at JILA since they need enhanced and controlled VUV light for applications in time- and angle-resolved photoelectron spectroscopy (trARPES). Specifically, we will explore energy scaling in our setup to see if we can increase the pulse energies. One strategy to use is to inject temporally stretched pulses into the hollow PCF waveguide. The success here depends on whether the damage mechanism for the PCF waveguide is primarily driven by the peak power of the laser, or the average power; i.e. is the delicate structure of the waveguide ablated, or melted? Experiments to-date are inconclusive in this regard. However, we will be working to improve the heat dissipation in mounting of this PCF waveguide, which is currently mounted in a fused-silica capillary.

Another approach to increasing the peak pulse energy, that was attempted at KMLabs, was to revert to a simple capillary hollow-waveguide for this upconversion. In a series of tests we did, our finding was that the overall conversion efficiency for this four-wave mixing process could not be recovered simply by increasing the energy of pulses injected into the waveguide proportionate to the increased mode area. Thus, the use of these antiresonant hollow PCF waveguides seems to be a crucial enabler for the HCHG process.

The setup developed under SBIR support will continue to be used, with an increasing emphasis on the area where simulations seem to direct efforts: toward the generation of higher-energy photons. This is also an area of DOE strategic interest: KMLabs currently is building a system for Argonne National Lab to use for VUV photoionization mass spectrometry (PIMS), and is in active discussion with another DOE laboratory for a Hyperion VUV system for different applications in materials science.

c. What opportunities for training and professional development has the project provided?

CU/JILA student Jeremy Thurston and postdoc Dr. Tika Kafle greatly benefitted from collaborations associated with this project - their goal has been to optimize the laser setup for use in time-resolved ARPES experiments at JILA for which this Pharos laser will ultimately be used. They worked previously with Dr. Dan Hickstein, who now is part of a new startup company Octave Photonics, and with Grant Buckingham, who gained experience directing research and is now doing research in quantum computing, and with Liam Weiner, who is KMLabs laser commercial product manager. As is often the case in Phase I projects, we completed enough work to gain valuable experience—but just enough to gain experience with the experimental setup and not enough to provide a compelling case for a Phase II project.

d. How have the results been disseminated to communities of interest?

Daniel Hickstein presented a summary of KM Labs VUV capabilities in a research talk at CLEO (Conference on Lasers and Electro-Optics) on May 19, 2021. As mentioned previously, the next Hyperion VUV system has been assembled and is under testing at KMLabs, bound for Argonne National Lab. We are optimistic that the system after this will be going to Los Alamos. These two DOE customers are using the system for very different applications—PIMS/ molecular science, and ARPES—materials science. Success in these installs will provide powerful market validation.

III. PRODUCTS: (Mandatory if products exist)

a. Publications, conference papers, and presentations

Daniel Hickstein presented a summary of KMLabs VUV capabilities in a research talk at CLEO (Conference on Lasers and Electro-Optics) on May 19, 2021. The work resulting in the Hyperion VUV system was published in *Optica* [DE Couch, DD Hickstein, DG Winters, SJ Backus, MS Kirchner, SR Domingue, JJ Ramirez, CG Durfee, MM Murnane, and HC Kapteyn, "Ultrafast 1 MHz vacuum-ultraviolet source via highly cascaded harmonic generation in negative-curvature hollow-core fibers," Optica **7**(7), 832-837 (2020). dx.doi.org/10.1364/optica.395688]. However, this paper slightly predated the SBIR. No new publications have resulted from this project.

b. Website(s) or other Internet site(s)

Nothing to Report

c. Technologies or techniques

The method of combining 1st, 2nd, and 3rd harmonic beams into a flowing-gas fiber will likely be a core technique of this product (The Hyperion VUV) as well as future products.

d. Inventions, patent applications, and/or licenses

The base technology for this HCHG upconversion process is now patented:

S.Backus, H. Kapteyn, D. Winters, “Two-Color Wave-Mixing Upconversion in Structured Waveguides,” US Patent 11,209,717 granted Dec 28,2021. This patent predates the SBIR; however, SBIR funds did serve to further elucidate the key aspects of this technology, and the setup as illustrated in Figure 1 will serve to further our understanding of the process and how to best utilize it.

e. Other Products

Nothing to Report

IV. PARTICIPANTS & OTHER COLLABORATING ORGANIZATIONS: MANDATORY

a. Participants

1) Name: Dr. Grant Buckingham

2) Project Role: PI
3) Nearest person month worked: 3
4) Contribution to Project: Dr. Buckingham has performed fundamental research to optimize the system and done project management on the hands-on work performed.
5) Funding Support: Other support from other DOE SBIR/STTRS
6) Collaborated with individual in foreign country: No
7) Country(ies) of foreign collaborator: NA
8) Travelled to foreign country: No
9) If traveled to foreign country(ies), duration of stay: NA

1) Name: Dr. Henry Kapteyn
2) Project Role: Key Personnel
3) Nearest person month worked: 1
4) Contribution to Project: Project management and scientific insight
5) Funding Support: University of Colorado Boulder and other support from other DOE SBIR/STTRS
6) Collaborated with individual in foreign country: yes
7) Country(ies) of foreign collaborator: Spain, Belgium, Germany, Sweden, Italy, Switzerland, China, Austria, Netherlands, Taiwan, South Korea. U.K., Czech Republic, Mexico
8) Travelled to foreign country: Yes
9) If traveled to foreign country(ies), duration of stay: <1 month

1) Name: Dr. Dan Hickstein
2) Project Role: Key Personnel
3) Nearest person month worked: 1
4) Contribution to Project: Project management and scientific insight
5) Funding Support: other support from other DOE SBIR/STTRS
6) Collaborated with individual in foreign country: No
7) Country(ies) of foreign collaborator: NA
8) Travelled to foreign country: No
9) If traveled to foreign country(ies), duration of stay: NA

1) Name: Eric Rinard
2) Project Role: Engineer
3) Nearest person month worked: 1
4) Contribution to Project: Updating design of system
5) Funding Support: Product revenue
6) Collaborated with individual in foreign country: No
7) Country(ies) of foreign collaborator: NA
8) Travelled to foreign country: Yes
9) If traveled to foreign country(ies), duration of stay: < 1 month

1) Name: Dr. Margaret Murnane
2) Project Role: Key Personnel
3) Nearest person month worked: <1 Month
4) Contribution to Project: Project management and scientific insight
5) Funding Support: University of Colorado Boulder
6) Collaborated with individual in foreign country: yes
7) Country(ies) of foreign collaborator: Spain, Belgium, Germany, Sweden, Italy, Switzerland, China, Austria, Netherlands, Taiwan, South Korea. U.K., Czech Republic, Mexico
8) Travelled to foreign country: yes
9) If traveled to foreign country(ies), duration of stay: 2 months

1) Name: Dr. Tika Kafle
2) Project Role: Postdoctoral Researcher
3) Nearest person month worked: 2
4) Contribution to Project: Dr. Kafle has worked on aligning the system with the new drive laser
5) Funding Support: University of Colorado Boulder
6) Collaborated with individual in foreign country: No
7) Country(ies) of foreign collaborator: NA
8) Travelled to foreign country: No
9) If traveled to foreign country(ies), duration of stay: NA

1) Name: Jeremy Thurston
2) Project Role: Graduate Student
3) Nearest person month worked: 1
4) Contribution to Project: Mr. Thurston has worked on aligning the system with the new drive laser
5) Funding Support: University of Colorado Boulder
6) Collaborated with individual in foreign country: No
7) Country(ies) of foreign collaborator: NA
8) Travelled to foreign country: No
9) If traveled to foreign country(ies), duration of stay: NA

b. Partners

Provide the following information for each partnership:

1. Organization Name: University of Colorado Boulder
2. Location of Organization: Boulder, Colorado
3. Partner’s contribution to the project: Collaborative research
4. More detail on partner and contribution: domestic

c. Other Collaborators

Nothing to Report

V. IMPACT (Optional but strongly Recommended)

a. What was the impact on the development of the principal discipline(s) of the project?

We believe this technology will allow us to offer a strong new line of products that produce high-power beams of VUV light, which is currently quite challenging for the laser community to product reproducibly.

b. What was the impact on other disciplines?

We are still learning of new fields that may be interested VUV generation. Angle-resolved photoemission spectroscopy (ARPES) makes use of this frequency range, as does a burgeoning field of chemical physicists and engineers studying combustion processes on the molecular level using photoionization mass spectrometry (PIMS). In general, the physical chemistry community has an unmet need for stable commercial products that produce VUV light.

c. What was the impact on the development of human resources?

Experience gained by Liam Weiner, who is now Hyperion product manager. This project also benefitted PhD and postdoctoral training through collaboration—primarily PhD student Jeremy Thurston and one postdoc Dr. Tika Kafle.

d. What was the impact on physical, institutional, and information resources that form infrastructure?

Nothing to Report

e. What was the impact on technology transfer?

This is a patented technology as described above, which predates this SBIR

f. What was the impact on society beyond science and technology?

The technology investigated here is an enabling tool, thus it quite possibly will be instrumental in development of new energy technologies.

g. What percentage of the award's budget was spent in foreign country(ies)?

None

VI. CHANGES/PROBLEMS: Optional (but strongly encouraged); Carryover Amount Mandatory

a. Changes in approach and reasons for change

We had two changes of PI during this project. Original PI Dr. Daniel Hickstein left to join a startup company, Octave Photonics. The second PI, Dr. Grant Buckingham, left to join Quantinuum. The job market in this photonics area is highly competitive.

b. Actual or anticipated problems or delays and actions or plans to resolve them

We needed to change to a new IR drive laser for this system and that caused delays. We also had delays as a result of COVID-19, because we had to reduce office/lab occupancy and we experienced supply chain delays.

c. Changes that have a significant impact on expenditures

Nothing to Report

d. Significant changes in use or care of human subjects, vertebrate animals, biohazards, and/or select agents

Not applicable.

e. Change of primary performance site location from that originally proposed

None.

f. Carryover Amount

This is the final project report. All funds are expended.

VII. DEMOGRACHIC INFORMATION: Mandatory (provide an email address for each participant)

GBuckingham@KMLabs.com; HKapteyn@KMLabs.com; DanHickstein@KMLabs.com; ERinard@kmlabs.com, Tika.Kafle@colorado.edu; Jeremy.Thurston@colorado.edu; Margaret.Murnane@colorado.edu

VIII. SPECIAL REPORTING REQUIREMENTS: Mandatory

Not applicable.

References

1. DE Couch, DD Hickstein, DG Winters, SJ Backus, MS Kirchner, SR Domingue, JJ Ramirez, CG Durfee, MM Murnane, and HC Kapteyn, "Ultrafast 1 MHz vacuum-ultraviolet source via highly cascaded harmonic generation in negative-curvature hollow-core fibers," Optica **7**(7), 832-837 (2020). dx.doi.org/10.1364/optica.395688

2. G Ycas, D Maser, and DD Hickstein, PyNLO—Python Package for Nonlinear Optics (GitHub Repository, 2018), https://Github.Com/PyNLO/PyNLO.

3. J Hult, "A Fourth-Order Runge–Kutta in the Interaction Picture Method for Simulating Supercontinuum Generation in Optical Fibers," J. Lightwave Technol. 25, 3770-3775 (2007) https://www.osapublishing.org/jlt/abstract.cfm?uri=jlt-25-12-3770