Advanced Photon Acceleration Schemes for Tunable XUV/Soft X-Ray Sources

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We report on the advances made under the purview of DOE award number DE-SC0019135 that was active during the period: 09/01/2018–08/31/2021. The grant investigated the application of a "flying focus" to the problem of photon acceleration—in which a dynamic refractive index gradient is used to continuously upshift a probe beam's frequency. Codes were written to describe the creation of ionization waves of arbitrary velocity (IWAVs) for use as a photon-accelerating medium, as well as the behavior of a witness pulse residing in said medium. Experiments first verified the spatiotemporal control over laser intensity provided by a chromatic flying focus, then used that ability to produce small-diameter IWAVs in the far-field with the expected dynamics, and finally demonstrated even further flexibility by producing large-diameter IWAVs in the laser quasi-far- field that maintained the beneficial dynamics. Multiple innovative diagnostics—spectrally resolved Schlieren and spectrally resolved interferometry—were pioneered in order to diagnose the IWAVs. For IWAV production in the laboratory, however, beam quality was identified as a key limitation in the quasi-far-field. Since the original chromatic flying focus was found to result in relatively long (ps duration) intensity peaks, which could limit some applications including photon acceleration, additional techniques were invented to provide similar spatiotemporal control while also retaining ultrashort intensity peaks. While simulations have identified several interesting regimes for photon acceleration—first predicting the upshift of a counterpropagating witness pulse from the optical to the extreme ultraviolet in less than 1 cm, and later obtaining similar shifts in less than 100 µm in a simpler self-seeded configuration—experimental demonstration is left for future work.

The personnel supported under this grant were D. Turnbull (P.I.), P. Franke, J. Palastro, A. Howard, D. Ramsey, T. Simpson, and D. Froula. Nine papers were published in various journals, including three in Physical Review Letters, and three invited talks were given at the EPS Conference on Plasma Physics (two) and the Anomalous Absorption Conference (one). The list of papers and invited talks is shown below.

Published manuscripts:

- J. Palastro, D. Turnbull, S.-W. Bahk, R. K. Follett, J. L. Shaw, D. Haberberger, J. Bromage, and D. H. Froula, "Ionization waves of arbitrary velocity driven by a flying focus," Phys. Rev. A **97**, 033838 (2018).
- D. Turnbull, P. Franke, J. Katz, J. P. Palastro, I. A. Begishev, R. Boni, J. Bromage, A. L. Milder, J. L. Shaw, and D. H. Froula, "Ionization waves of arbitrary velocity," Phys. Rev. Lett. **120**, 125001 (2018).
- A. Howard, D. Turnbull, A. S. Davies, P. Franke, D. H. Froula, and J. P. Palastro, "Photon acceleration in a flying focus," Phys. Rev. Lett. **123**, 124801 (2019).
- P. Franke, D. Turnbull, J. Katz, J. P. Palastro, I. A. Begishev, J. Bromage, J. L. Shaw, R. Boni, and D. H. Froula, "Measurement and control of large diameter ionization waves of arbitrary velocity," Opt. Exp. **27**, 31978 (2019).
- D. Turnbull, S.-W. Bahk, I. A. Begishev, R. Boni, J. Bromage, S. Bucht, A. Davies, P. Franke, D. Haberberger, J. Katz, T. J. Kessler, A. L. Milder, J. P. Palastro, J. L. Shaw, and D. H. Froula, "Flying focusing and its application to plasma-based laser amplifiers," Plas. Phys. & Cont. Fus. **61**, 014022 (2019).
- D. Froula, J. P. Palastro, D. Turnbull, A. Davies, L. Nguyen, A. Howard, D. Ramsey, P. Franke, S.-W. Bahk, I. A. Begishev, R. Boni, J. Bromage, S. Bucht, R. K. Follett, D. Haberberger, G. W. Jenkins, J.

Katz, T. J. Kessler, J. L. Shaw, and J. Vieira, “Flying focus: spatial and temporal control of intensity for laser-based applications,” Phys. Plasmas **26**, 032109 (2019).

- P. Franke, D. Ramsey, T. T. Simpson, D. Turnbull, D. H. Froula, and J. P. Palastro, “Optical shock-enhanced self-photon acceleration,” Phys. Rev. A **104**, 043520 (2021).
- D. Ramsey, B. Malaca, A. Di Piazza, M. Formanek, P. Franke, D. H. Froula, M. Pardal, T. T. Simpson, J. Vieira, K. Weichmann, and J. P. Palastro, “Nonlinear Thomson scattering with ponderomotive control,” Phys. Rev. E **105**, 065201 (2022).
- D. Ramsey, A. Di Piazza, M. Formanek, P. Franke, D. H. Froula, B. Malaca, W. B. Mori, J. R. Pierce, T. T. Simpson, J. Vieira, M. Vranic, K. Weichmann, and J. P. Palastro, “Exact solutions for the electromagnetic fields of a flying focus,” Phys. Rev. A **107**, 013513 (2023).

Invited talks:

- D. Turnbull, “Flying focus and its application to plasma-based laser amplifiers,” EPS Conference on Plasma Physics, Prague, CZ (2018).
- P. Franke, “Ionizations waves of arbitrary velocity in the quasi-far-field,” Anomalous Absorption Conference, Telluride, CO, USA (2019).
- P. Franke, “Spatiotemporal control of laser pulses for broadband extreme ultraviolet generation,” EPS Conference on Plasma Physics, virtual (2021).

I. MOTIVATION

The development of bright, high-quality light sources in the extreme ultraviolet (XUV) and x-ray spectral regions is expected to benefit many fields of science and technology, including nanoscience/nanotechnology, materials science, chemistry, biology, and physics. At the forefront of high energy density physics, x rays are being used to probe extreme states of matter in order to understand material properties in environments ranging from planetary cores to the stagnation stage of inertial confinement fusion implosions. Creating such probes from ``table-top'' sources would dramatically enhance their ubiquity. A number of schemes have been proposed, many of which are plasma-based—exploiting the fact that plasmas are capable of withstanding extremely high electric and magnetic fields without breakdown, and can be regenerated at negligible cost and high repetition rate. Current approaches, however, typically suffer from: poor tunability, incoherence, polychromaticity, large divergence, low efficiency, lack of polarization control, and the need for a high intensity laser.

Photon acceleration can be used to frequency-shift a conventional optical light source into the XUV/x-ray range while retaining the coherence, divergence, monochromaticity, and even polarization of the original source. In a photon accelerator, a time-varying refractive index is used to change the frequency of photons passing through it [1]. Since the refractive index of plasma is proportional to its density, a steep plasma density gradient can be used *if it can be synchronized with the propagation of the upshifting light source*. The energy efficiency of a photon accelerator is inversely proportional to the frequency upshift, but this can be several orders of magnitude higher than other schemes.

A laser-produced ionization wave is an attractive medium for photon acceleration [2-6]. Frequency shifts accumulate during the time a photon spends within the refractive index gradient, and in this respect laser-produced fronts are ostensibly ideal because they move close to the speed of light (Fig. 1). Nevertheless, ``phase slippage'' between the ionization front and the upshifting probe beam has been a key limitation because the group velocity of the probe accelerates as its frequency increases such that it decouples from the gradient region. On the one hand, high plasma densities are desirable to produce stronger refractive index gradients and associated shifts; on the other hand, the high density leads to low ionization-wave velocities, exacerbating the dephasing. Dense plasmas also strain the ability to generate ionization over long distances because the plasma tends to both refract and absorb the conventionally-focused ionizing laser, limiting further propagation.

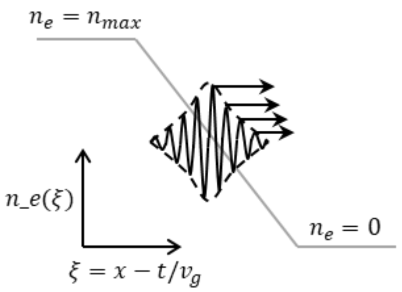

Figure 1. Cartoon of a laser pulse envelope co-propagating with a plasma density gradient in the moving frame of the ionization wave. The laser will frequency upshift as it propagates.

II. PROGRESS UNDER THIS GRANT

Under this grant, the ``flying focus'' was brought to bear on the problem of photon acceleration in order to circumvent the issues previously identified. Flying focus is now something of an umbrella term that refers to various techniques providing spatiotemporal control over the velocity of a laser intensity peak—decoupling it from the group velocity of the laser itself. The original concept combined linear chirp with chromatic aberration to achieve such control [7,8]. By stretching a laser pulse in time using a grating pair and then focusing it with a diffractive optic designed to focus different colors within the pulse to different locations along the laser axis, an extended focal region is created that can be much longer than the Rayleigh length, within which the peak intensity can propagate at any velocity. Three examples are shown in Figure 2, where simply changing the laser chirp modifies the focal speed to be (a) forward subluminal, (b) backward luminal, or (c) infinite. When the flying focus was first conceived, we immediately thought to use it to create ionization waves of arbitary velocity (IWAVs) to advance applications like photon acceleration and Raman amplification [9,10].

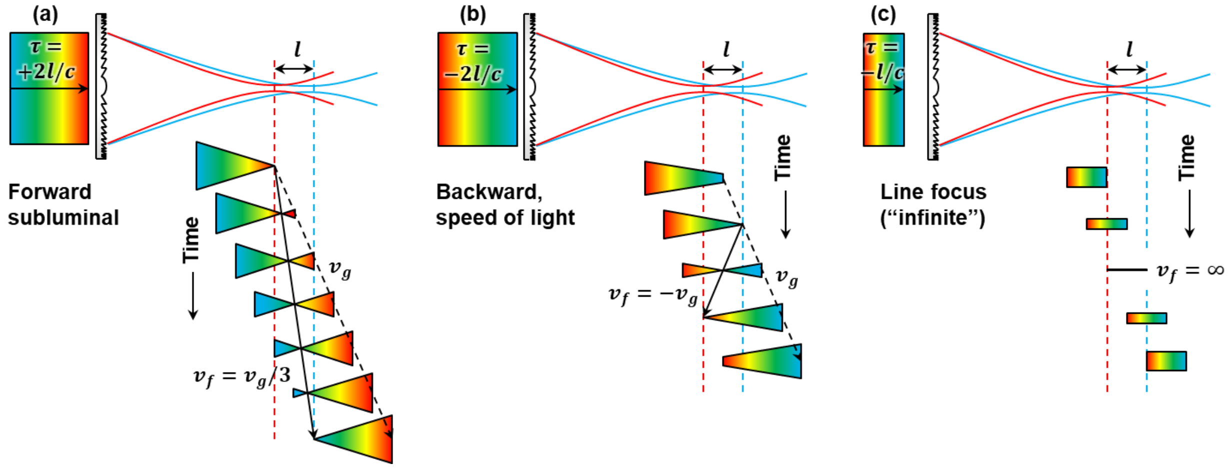

Figure 2. Examples of the flexibility in tuning the focal-spot velocity using the chromatic flying focus, ranging from (a) forward subluminal, to (b) backward luminal, and (c) infinite.

A laser propagation code was written to self-consistently model the propagation of the chromatic flying focus pulse, ionization dynamics of a background medium, and the resulting plasma refraction and depletion of laser energy. The simulations confirmed that IWAVs can be formed and that the laser can maintain self-similar propagation throughout the focal region in spite of the ionization dynamics [11]. They also found that backward and superluminal IWAV propagation mitigate the issue of ionization-induced refraction that typically affects conventionally focused laser pulses (as well as subluminal forward-propagating flying focus beams).

We immediately set out to validate those predictions in the laboratory using the Multi-Terawatt (MTW) Laser at the University of Rochester Laboratory for Laser Energetics [12]. The first experiments used just ~25 mJ to form a narrow (~10 μm) plasma channel in atmospheric air at best focus. The experimental setup is shown in Fig. 3(a), with examples of the data in Fig. 3(b). The 8.7-nm-FWHM-bandwidth pulse (with a flat power spectrum) was stretched anywhere from ~500 fs to ~40 ps with either positive or negative chirp. It was then focused by a diffractive lens with a 51-cm focal length, yielding an extended focal region more than 4 mm in length. A portion of this beam was split off, frequency doubled, optically delayed, and then used to illuminate the entire extended focal region in a side-on configuration relative to the primary flying focus beam. The focal region was imaged onto the entrance slit of a spectrometer-CCD pair with a Schlieren stop along the imaging path, which ensured that only light refracting off the edge of the plasma channel would reach the detector. A key innovation was the use of a chirped probe beam to provide sub-ps time resolution without the actual use of any ultrafast detectors. Since the probe wavelength was directly related to time, the wavelength first appearing at each axial location indicated the time at which plasma was first formed there, such that the time-integrated images captured the full IWAV dynamics. The main result was that we confirmed nearly ideal IWAV propagation using the chromatic flying focus [13].

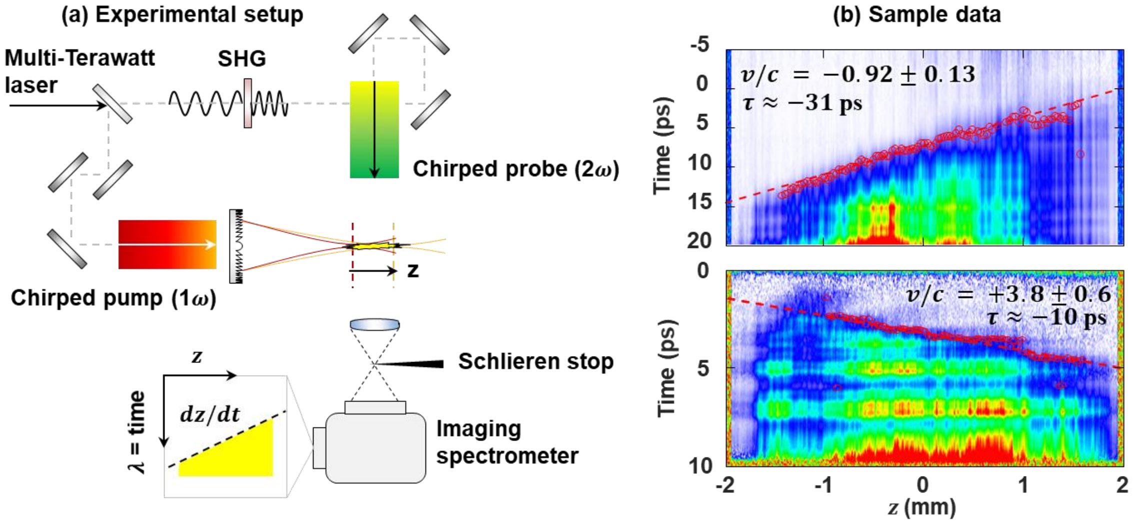

Figure 3. (a) Experimental setup used to diagnose ionization waves of arbitrary velocity (IWAVs) using the MTW laser. (b) Examples of the IWAV data.

The plasmas, however, were very narrow, and some applications (including photon acceleration) could benefit from having the same dynamics but in a wider channel. We therefore attempted to extend the scheme's flexibility by operating in the quasi-far-field with higher laser energy. The basic idea is that ionization is expected to follow the intensity isosurface corresponding to the ionization threshold in the medium. In the first experiment (at low energy), that intensity threshold was reached in the far-field at best focus. With additional energy, the isosurface moves towards the focusing lens into the quasi-far-field. This was found to be feasible both in simulations and in experiments by increasing the primary beam energy up to 5 J, forming channels more than 100 µm in diameter [14]. The channels were diagnosed using chirped interferometry, which provided all of the same information regarding the ionization dynamics while also yielding quantitative information about the plasma density.

This second experiment identified some limitations, however. Further amplifying the MTW laser produced a gain-narrowed power spectrum that was not nearly as flat. With varying amounts of power in each color, each wavelength reached the ionization threshold at positions that did not vary linearly with respect to one another, yielding a dynamic (nonconstant velocity) IWAV trajectory [14]. While in principle perfect control over the spectrally dependent laser power could make this a useful feature for obtaining arbitrary dynamics of the peak intensity, in practice this places very stringent requirements on the drive laser. In a similar vein, laser focal spot quality tends to degrade away from the far-field, so the larger channels were not as uniform. One might think to introduce a phase plate in order to restore operation to the far-field with a larger spot, but this would not be useful because ionization would be triggered upstream as soon as the hottest speckles reached the ionization threshold. Such limitations must be kept in mind in any scheme seeking to use wide channels produced by a flying focus.

Considering the challenges such as these anticipated for an experimental demonstration of photon acceleration, any such attempt should be well informed by simulations to identify the most promising conditions. To predict the performance of a photon accelerator using a flying-focus-produced IWAV, a 1-D photon kinetics code was coupled to the 2-D (cylindrically symmetric) laser propagation code described earlier. Special care was taken to ensure the validity of the 1-D assumption. Noting that the plasma channels produced in uniform gas by a Gaussian (TEM_{00}) laser pulse were centrally peaked, which implies the plasma would act as a negative lens and refract light out of the channel, a parabolic profile was instead used as the initial condition of the gas such that it would ionize into a guiding profile. As shown in Fig. 4, the photon kinetics simulations then predicted than an optical witness pulse injected into the backward-propagating (relative to the flying focus drive pulse) IWAV traveling at the speed of light in vacuum would be upshifted more than 4x to below 100 nm over a propagation distance of ~1 cm [15]. It was noted that obtaining larger frequency upshifts would require either more bandwidth and/or steeper density gradients, and that the sharpness of the density gradient is limited by the longer-than-transform-limited effective pulse duration of the intensity peak that is characteristic of the chromatic flying focus [16].

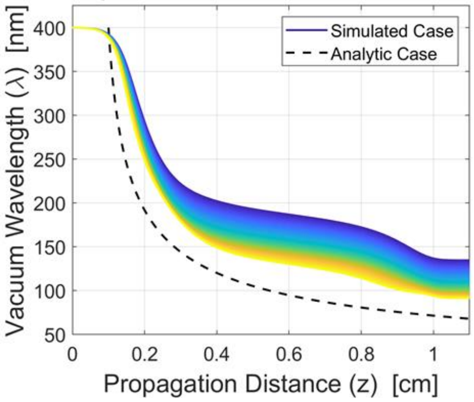

Figure 4. Simulations suggest a photon accelerator using the flying focus could upshift an optical laser by 4x within just 1 cm.

Since the relatively long effective pulse duration could also limit the utility of the flying focus for other applications, we started to investigate alternative flying focus approaches that might have the same beneficial dynamics without the same limitations, and several good candidates were identified. For example, the combination of radial group delay (provided, e.g., by a radial echelon optic) and spherical aberration (from an axiparabola) results in a tunable-velocity intensity peak that retains an ultrashort duration [17]. Temporal pulse shaping combined with nonlinear self-focusing provides yet another possible solution [18], and most recently it was also noted that temporal pulse shaping of a stencil pulse can control the focusing dynamics of a separate witness pulse through cross-phase modulation in a Kerr lens [19].

Further exploring the large possible photon acceleration parameter with simulations, another even more promising approach was discovered. Using a flying focus technique such as those just described that retains an ultrashort pulse duration, a transverse intensity profile that is peaked off-axis to provide self-guiding due to the combination of two orthogonally polarized Laguerre-Gaussian modes, and a preformed partially ionized (Z=3) nitrogen plasma, simulations predict that photons in the flying focus pulse can be trapped and upshifted almost an order of magnitude within less than 100 μm while also getting compressed to a shorter duration, at a relatively high energy efficiency of 0.12%—more than 100x higher than the previous results [20]. While this would still be a challenging experimental setup, it is certainly not infeasible, and the resulting probe would be well worth the effort.

III. CONCLUSIONS

In summary, over the duration of the grant, significant progress was made toward the goal of demonstrating a photon accelerator. Simulations identified several promising setups utilizing a flying focus to drive an ionization front that would result in a signal in the extreme ultraviolet range. Experiments demonstrated the ability to produce IWAVs—a key ingredient. Chirped Schlieren and chirped interferometry diagnostics were developed to diagnose the sub-ps dynamics of the plasma channel formation without the actual use of any ultrafast detectors. Although an experimental demonstration of efficient photon acceleration into the XUV range is left for future work, we have enough information to conclude that it should certainly be technically feasible.

IV. ACKNOWLEDGMENTS

This final report summarizes the work supported by the DOE FES under Award Number DE-NA0019135. This report was prepared as an account of work sponsored by an agency of the U.S. Government. Neither the U.S. Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof.

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