Diagnostic of Laser-Plasmas: Single-shot Supercontinuum Spectral Interferometry

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Abstract. Ultrafast optical diagnostics play a vital role in probing the dynamics in laser-matter interactions, including those observed in the high intensity ultrashort laser pulse regime. We developed a new femtosecond optical diagnostic, single-shot supercontinuum spectral interferometry (SSSI), which measures ultra-rapid transients in the complex index of refraction induced by an intense laser pulse. This measurement provides a direct view on how the laser-produced perturbations evolve in time and space. To date, SSSI has been successfully used to diagnose femtosecond dynamics in the interaction of intense laser pulses with gases, nanometer-sized atomic or molecular clusters, and plasmas, including plasma waveguides.

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INTRODUCTION

Frequency domain or spectral interferometry (SI) has been employed in many experiments to measure refractive index transients. These include measurements of self-phase modulation in optical fibers, induced phase modulation in solids, time evolution of femtosecond laser-plasmas [1], and laser-driven plasma wakefields [2]. In this technique, a reference pulse and a time delayed probe pulse, upon which a pump-pulse-induced phase has been imposed, interfere in the frequency domain when combined in a spectrometer [1,2]. Recently, single-shot spectral interferometry (SSI) for the measurement of transient refractive index variations was realized by linearly chirping the reference and probe beams so that each temporal slice of the refractive index variation was projected onto a different frequency component [3,4]. The temporal phase variation was then obtained by applying a direct map from frequency to time. However, aside from resolution limits imposed by the spectrometer, this method has a fundamental limitation determined by the spectral bandwidth and the degree of chirp.

Here, we demonstrate a SSI diagnostic that uses chirped supercontinuum (SC) pulses generated in air. We call this technique single-shot supercontinuum spectral interferometry (SSSI) diagnostic [5]. Using SSSI, we can achieve single-shot temporal resolution (~10 fs) up to an order of magnitude better than in previous work [3,4,6].

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The use of air as the nonlinear medium provides, in a single self-focused filament, probe light almost three orders of magnitude brighter than the SC generated in solids. We note that the proximity of the SC spectrum to the pump wavelength reduces group velocity walk-off effects, which can be present in spectral interferometry schemes using frequency doubled probe pulses [7]. In addition, the temporal field of view can be arbitrarily adjusted by chirping the probe beam with an appropriate thickness of dispersive material, which is easier than pulse stretching by limiting the phase-matching bandwidth [7]. The very large bandwidth of our SSI probe pulses demands that a detailed analysis be performed in order to determine the method’s ultimate temporal resolution. To this end, we have examined, experimentally and theoretically, the dependence of the temporal resolution of this diagnostic on the SC pulse bandwidth and chirp [5].

ULTRAFAST OPTICAL DIAGNOSTIC

Our SSSI layout is shown in Fig. 1. The SC pulse is generated as broadband conical emission from f/8 focusing of a ~1 mJ, 80 fs pulse in 1 atm of air. Although it is possible to generate SC in a sapphire window, the pump power is limited to the critical power for self-focusing ($P_{cr} \approx 3$ MW) to avoid multiple filament formation, which leads to significant spatio-temporal phase distortions. This limits maximum useable pump energies in bulk media to the µJ range. The SC beam is collimated and split into collinear twin pulses (reference and probe pulses) with delay $\tau$ by a Michelson-interferometer-type delay line. The twin pulse beam is sent through a ~250 µm diameter aperture to clean the spatial profile and reduce the spatial chirp. After that, temporal chirp was added with a dispersive glass window (1” thick SF4). Then, the twin chirped SC pulses were directed through a pump mirror (dichroic splitter), and collinearly focused with a pump beam to a FWHM spot size of ~170 µm, overfilling the pump spot. The pulse timing was arranged so that the reference pulse preceded the probe pulse, and the pump pulse was overlapped with the probe, generating a time- and space-dependent phase variation. After the interaction, the pump was removed from the beam path by a dichroic mirror. The twin pulses were then imaged with 15X magnification onto a spectrometer slit, producing a spectral interferogram on the spectrometer’s focal plane CCD with one-dimensional (1D) space resolution along the slit. The inset in Fig. 1 shows a sample of spectral interferograms.

As a test of the diagnostic, we have measured optical Kerr-induced cross-phase modulation (XPM) in a fused silica glass. Figure 2(a) shows a 1D space-resolved XPM profile extracted from a spectral interferogram with the frequency-to-time direct mapping technique. It shows that the profile does not fully reproduce the Gaussian-like temporal structure of the pump pulse. This is attributed to the fundamental limitation of the direct mapping technique, where the temporal resolution is related to the degree of chirp and spectral bandwidth [5]: For a given finite spectral bandwidth, the large chirp necessary to broaden the observation window can dramatically degrade the temporal resolution. To overcome this limitation, a Fourier transform method was developed, the full details of which are described in ref [5]. Using this technique, a complete phase shift profile was reconstructed and is shown in Fig. 2(b); evidently, the
distortion associated with direct mapping is greatly reduced. In conclusion, our SSSI diagnostic can measure laser-induced ultrafast refractive index transients, providing ~10 fs resolution and a ~1 ps observation window with single-shot operation.

APPLICATIONS IN LASER-PLASMA INTERACTIONS

Using SSSI, we have investigated the interaction of intense laser fields with various target media–gases, atomic clusters, and preformed plasma waveguides. Many interesting ultrafast phenomena in these interactions are described as follows.

Laser-Gas Interaction

The interaction of high-intensity ultrashort pulses with gases is rich in fundamental phenomena and applications including optical field ionization (OFI), high harmonic
generation, relativistic channeling, and laser-induced wake-fields in plasmas. In particular, OFI is a fundamental and universal process that occurs in a wide range of media under high-intensity femtosecond laser irradiation $I > 10^{15}$ W/cm$^2$. Here, we use SSSI to characterize OFI of low-Z gases such as helium.

The experimental setup is shown in Fig. 1, where the target is now a helium gas jet. A 20 mJ, 240 fs pump pulse is focused into the helium gas jet with a peak intensity $I_{\text{peak}} = 3.8 \times 10^{16}$ W/cm$^2$. Figure 3 (a) shows the spatio-temporal phase shift profile extracted from a typical spectral interferogram at 15-psi jet backing pressure. Here, the phase shift is attributed to the density of free electrons liberated from helium atoms by the laser field. Hence, the central line-out shown in Fig. 3 (b) shows the signature of temporal double-step ionization of helium (He $\rightarrow$ He$^+$ $\rightarrow$ He$^{2+}$). This measurement of electron density evolution is in good agreement with the OFI model [8] except that the measured steps are less distinct than expected. The explanation is that an excessively long laser-gas interaction length can smear out any temporal features we wish to uncover [9]. In our experiment, the laser-target interaction length was limited to a minimal length of $\sim 0.5$ mm to observe these transients. In conclusion, we have measured time-resolved optical field ionization of helium using SSSI, confirming sequential ionization: He $\rightarrow$ He$^+$ followed by He$^+$ $\rightarrow$ He$^{2+}$.

**Laser-Cluster Interaction**

The interaction of intense laser pulses with atomic clusters, van der Waals-bonded aggregates of up to $\sim 10^7$ atoms, is of great current interest. The generation of x-rays, fast electrons and ions, and fusion products are some of the applications, all of which are strongly affected by the details of laser coupling to the resulting cluster plasma.

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**FIGURE 3.** (a) Experimental spatio-temporal phase profile from optical field ionization of helium at 15-psi jet backing pressure. (b) Central line-outs for jet backing pressure of 5 psi (line with solid triangles) and 15 psi (line with squares). The pump pulse envelope obtained from XPM in glass is also shown (line with circles). The inset shows spatial phase profiles from the 5-psi case at 20 fs increments.
Here, we elucidated cluster explosion dynamics by measuring the cluster transient complex polarizability $\gamma$ using SSSI [10]. The evolution of $\gamma$ both embodies the details of the exploding cluster dynamics and points the way to several applications, including the self-focusing effect demonstrated recently [11].

The experiment setup is same as shown in Fig. 1, where the target is replaced by a supersonic argon cluster gas jet. The mean cluster radius is estimated using the Hagenau parameter [12] to be in the range 150–300 Å. A 1 mJ, 800 nm, 80 fs pump pulse was focused into the supersonic gas jet collinearly with ~1.5 ps chirped SC reference and probe pulses of SSSI. Figure 4 shows the results of our SSSI measurements and calculation results from our laser-cluster interaction model [13]. The transient phase shifts $\Delta\phi(t)$ and small signal absorption coefficients $\eta(t)$ on the beam axis are plotted in Fig. 4(a) and (b), respectively. $\bar{\gamma}_r$ and $\bar{\gamma}_i$ are shown calculated in Fig. 4(c) and (d) for the same range of average cluster sizes. The ensemble average $\bar{\gamma}$ is calculated by determining $\gamma$ for a range of individual cluster sizes centered on the average, and then averaging over a 100% FWHM size distribution. The simulation results are in good agreement with the experimental results.

The interpretation of these results derives from detailed examination of our calculation results [13]: If the optical response of above-critical density layers inside an exploding cluster dominates that of the sub-critical layers outside, the real polarizability of the cluster is positive ($\gamma_r > 0$). However, as the cluster expands, the sub-critical density layers begin to dominate the optical response and the real

![FIGURE 4.](image_url)

FIGURE 4. (a) Phase shift (left scale) and corresponding real index shift (right scale) extracted from spectral interferograms, for backing pressures (cluster radii) 150 psi (150 Å), 200 psi (200 Å), 250 psi (235 Å), 300 psi (270 Å), and 350 (300 Å). (b) Small signal absorption coefficient (left scale) and corresponding imaginary index (right scale). (c)-(d) Calculation of real and imaginary ensemble-averaged polarizability for average cluster sizes 150–300 Å.
polarizability goes negative ($\gamma_r < 0$). The peak of $\gamma_i$ occurs at the zero crossing of $\gamma_r$. The individual cluster dynamics translate to similar dynamics in the ensemble result for $\delta n_r$ and $n_i$. This behaviour is consistent with the heated cluster exploding layer-by-layer, with a coupling resonance occurring at the critical density surface of the expanding plasma [13]. This transient evolution of $\bar{\gamma}$ has a remarkable macroscopic effect – self-focusing of the pulse [11]. As a single cluster is heated and expands, $\gamma_r$ starts positive and grows with time. This occurs faster on the laser beam axis, where the intensity is higher, than at the beam edge. Hence, for sufficiently short pulses, an ensemble of laser-heated clusters provides a refractive index structure suitable for beam self-lensing.

**Laser-Plasma Waveguide Interaction**

Plasma waveguides are essential for optical guiding of intense laser beams over distances greatly in excess of the Rayleigh length for many applications, which include extremely high harmonic generation, x-ray lasers, and laser-plasma-based charged particle accelerators. These applications would benefit greatly from a large intensity-interaction length. Hence, it is worthwhile to understand how intense laser pulses interact with a plasma waveguide, in particular when they are injected and guided into the waveguide.

Here, we used our SSSI diagnostic to measure ultrafast modulation induced by intense pump pulses injected into a plasma waveguide. Figure 5(a) shows a scheme for generating a plasma waveguide and subsequent guiding of intense Ti:sapphire laser pulses, along with SSSI diagnostic beams. A 1064 nm, 100 ps, ~500 mJ laser pulse from a Nd:YAG laser system generated the plasma waveguide at the ~1cm long line focus of a $30^\circ$ base angle axicon in a static-filled gas of 640 torr of helium plus 10 torr of $N_2O$, with a peak on-axis intensity of $5 \times 10^{13}$ W/cm$^2$. A ~40 mJ, 70 fs pulse from our Ti:sapphire laser system was injected into the plasma waveguide with a peak intensity of $\sim 10^{17}$ W/cm$^2$.

![FIGURE 5. (a) Schematic of waveguide generation and intense pulse injection and guiding. (b) Extracted transient phase shift induced by an intense laser pulse injected into the plasma waveguide.](image-url)
Figure 5(b) shows the extracted transient phase shift $\Delta \Phi(x, t)$ imposed on the SC probe pulse, where $x$ is a coordinate transverse to the guide. We attribute this phase shift to pump-induced ionization at the waveguide entrance (independent interferometric measurements show that the waveguide is fully ionized beyond the entrance). This is suggested by the negative sign of the phase shift, which corresponds to ionization, and by the temporal location of the shift beginning near the center of the chirped SC pulse time window, where the pump pulse is located.

CONCLUSIONS

We have developed a novel ultrafast optical diagnostic and used it to investigate femtosecond time-resolved dynamics of intense, ultrashort laser interaction with various targets – gases, nanometer-sized clusters, and plasma waveguides. So far, we have successfully observed, using the SSSI diagnostic, (i) the laser-induced double stepwise optical ionization of helium, (ii) time-resolved explosion dynamics of intense-laser-heated atomic clusters, and (iii) the coupling and guiding of intense laser pulses injected into a plasma waveguide.

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