

Self-Compression of Ultrashort Pulses through Ionization-Induced Spatiotemporal Reshaping

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We present the first demonstration of a new mechanism for temporal compression of ultrashort light pulses that operates at high (i.e., ionizing) intensities. By propagating pulses inside a hollow waveguide filled with low-pressure argon gas, we demonstrate a self-compression from 30 to 13 fs, without the need for any external dispersion compensation. Theoretical models show that 3D spatiotemporal reshaping of the pulse due to a combination of ionization-induced spectral broadening, plasma-induced refraction, and guiding in the hollow waveguide are necessary to explain the compression mechanism.

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The propagation of intense femtosecond duration light pulses in materials, gases, and plasmas is important for many areas of high field science, such as high-harmonic generation [1,2], laser-particle acceleration [3], the study of plasma and nonlinear dynamics [4], and light detection and ranging based on atmospheric propagation of laser beams [5]. In past work, nonlinear propagation has also been employed extensively in ultrafast technology for temporal compression of pulses to an even shorter duration. Nonlinear self-phase modulation (SPM) experienced by a pulse as it travels through a medium will spectrally broaden it. This broadened pulse bandwidth can then be compressed by applying appropriate dispersion to the pulse after the spectral broadening. Typically, a pulse is self-phase modulated by propagation through an optical fiber [6,7] or a gas-filled hollow optical waveguide [8], and dispersion compensation is provided by pairs of prisms or gratings. These schemes have been employed to compress pulses to <4 fs pulse duration [9]; however, they are also relatively complex and lossy. Furthermore, all previously demonstrated schemes are limited to nanojoule to submillijoule energies, by waveguide propagation, and damage of the nonlinear material. SPM-based techniques also tend to generate pulses with large spectral modulations and complex nonlinear chirps that are difficult to compensate for, resulting in large temporal pedestals on the pulse. A variety of other schemes for pulse compression have been demonstrated or proposed, for example, phase modulation of a pulse induced by molecular phase wave packets [10–12] using vibrational or rotational motion. However, these schemes are also limited to low laser intensities below the dissociation threshold of the molecule. The use of ionization to blueshift and to phase modulate a pulse for compression has been previously considered [13–15]; however, this scheme also depends on the use of external dispersion compensation and is distinct from the mechanism seen in this work. Finally, self-compression of weakly relativistic intensity laser pulses in plasmas using 1D and 3D simulations was recently proposed theoretically [4]. That scheme required

a difficult-to-realize multilayer plasma geometry to avoid plasma instabilities.

In this work, we discovered a new and unanticipated regime of ultrashort pulse compression that operates at high laser intensities, where the pulse is ionizing the medium and where 3D effects dominate the compression mechanism. We show that an intense 800 nm wavelength pulse propagating through a short gas-filled hollow waveguide, can reshape and *self-compress* in time without the need for subsequent dispersion compensation. Theoretical models show a new mechanism for pulse compression that cannot be explained by 1D models, where 3D spatiotemporal reshaping of the pulse occurs by ionization-induced spectral broadening, plasma-induced refraction, and guiding in the hollow waveguide. Therefore, this new technique is fundamentally different from previous mechanisms that broaden and compress in separate steps. This high field pulse compression is unique in its ability to shorten the duration of intense light pulses with high efficiency and with excellent pulse fidelity. Because this new mechanism can operate at intensities that are significantly higher than in other schemes, it may be scalable to much higher pulse energies, with implications for the design of many high field science experiments. Finally, our work is the first to demonstrate both experimentally and theoretically that it is possible to take advantage of 3D pulse evolution to compress a light pulse in a way that is not predicted in 1D simulations. Thus, our work provides unique insight that fundamentally new physical phenomena can be found using 3D simulations that has important implications for understanding many high field and plasma science experiments using either plasma or fiber-based guided geometries.

In our experiment, pulses from a Ti:sapphire amplifier with ≈ 30 fs near-transform-limited pulse duration and with energy of 2.2 mJ, at a repetition rate of 1 kHz [16], were focused into a 2.5 cm long, 150 μm inner diameter, hollow waveguide filled with low-pressure argon. The spot diameter of the laser at the entrance to the waveguide was 50 μm FWHM. The energy throughput of the wave-

guide is $\sim 60\%$ with no gas present, adjusted for other losses. The focusing region before and after the waveguide was held under vacuum to limit the interaction region and to maintain a constant pressure [17]. The pulse was characterized before and after the waveguide using second-harmonic frequency-resolved optical gating (SHG-FROG) [18]. All FROG measurements were deconvolved using commercial software from Femtosoft Technologies (Version 3.03) to an error of $<1\%$ for a 256×256 grid. After exiting the waveguide, the pulse passes through a $250 \mu\text{m}$ sapphire window and a 1 mm thick fused silica beam splitter in the FROG apparatus; the dispersion of these elements corresponds to an additional pulse chirp that stretches the measured pulse duration of at most 1–3 fs, and therefore does not affect our findings. Therefore, our pulse width measurements accurately determine the pulse duration at the exit of the waveguide. With no gas, we observed no change in the pulse after propagating through the waveguide.

The intensity in the waveguide, at approximately $10^{15} \text{ W cm}^{-2}$, is high enough that the argon gas is fully ionized on the leading edge of the pulse. This rapidly changing ionization during the leading edge of the pulse results in an index of refraction that decreases rapidly with time, causing an overall blueshift in the laser spectrum of tens of nanometers, which has been observed previously [19]. Figure 1 shows the pulse spectra at the exit of the waveguide with and without gas in the guide. Pressures as low as 1 torr result in a significant blueshift and broadening. As the pressure is increased, the pulse width also decreases to a minimum value of 13.3 fs at pressures around 4 torr. The compressed pulse width then remains relatively constant for increased pressures up to 9 torr, at which point the laser mode begins to break up. Both the input pulse and the compressed final pulse have a pulse width within 20% of the time-bandwidth product, as shown in Fig. 2. Figure 2(a) shows the temporal envelope of the pulse, both with and without gas in the

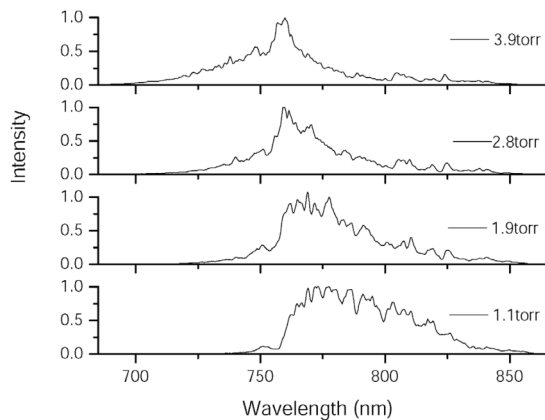


FIG. 1. Spectra of pulses emerging from waveguide filled with argon pressures of 4 torr decreasing to 1.1 torr. Ionization induces a spectral blueshift and broadening of the spectrum.

waveguide. The initial pulse width of 28.9 fs has been dramatically reduced to 13.3 fs at a pressure of 4 torr. The measured and deconvolved pulse spectra from the FROG are shown in Fig. 2(b) and correspond to a transform limit of 11 fs. The excellent agreement between the measured and retrieved spectra indicates that the measurement has high accuracy and is self-consistent, and that the output beam quality is high since the entire beam was characterized by the FROG. Moreover, the temporal phase varies by less than 0.4 rad over the duration of the pulse and small pedestals present on the pulse are in contrast to other mechanisms where nonlinear phase modulation and dispersion levels are high. This can lead to spectral components appearing at different times within the pulse, with uncompressible components that lead to large temporal wings on the pulse. At a pressure of 4 torr, the energy loss from the compression process is

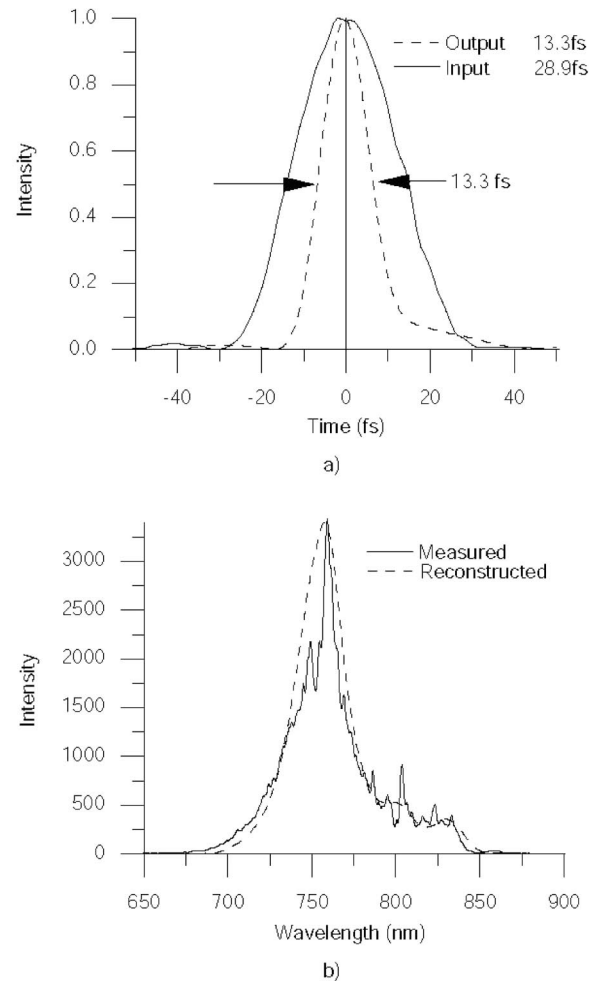


FIG. 2. Input and output pulse characteristics at the 4 torr optimum pressure for compression of pulse with an input laser intensity of $10^{15} \text{ W cm}^{-2}$. (a) Temporal profile of the input pulse (29 fs) and the final compressed pulse (13.3 fs). (b) Measured spectrum of the compressed pulse, compared with the retrieved spectrum from the FROG data. The excellent agreement confirms the fidelity of the pulse measurement.

only 5% of the total pulse energy, i.e., the output energy drops by 5% at 4 torr, while at 8 torr the loss is 10%. We observe lower levels of compression (from 30 to 24 fs) using a larger diameter, 230 μm , waveguide, and minimal amounts of compression by <1 fs when the waveguide is replaced by a large-diameter cell.

Conventional 1D pulse propagation models, such as those used to explain self-phase modulation in optical fibers, microstructured fibers, or hollow waveguides, fail to explain the observed compression phenomenon. In conventional pulse compression techniques, spectral broadening is accomplished through SPM. However, SPM alone broadens the pulse spectrum but does not alter the temporal envelope. Higher-order corrections due to self-steepening and other effects can reshape the trailing edge of the pulse but will not shorten the pulse in time without subsequent pulse compression. At high intensities, however, significant spectral and temporal effects are expected due to the rapid ionization on the leading edge of the pulse that leads to a rapidly decreasing plasma index $n_p = \sqrt{1 - (\omega_p^2/\omega^2)}$, where ω_p is the plasma frequency given by $\omega_p^2 = n_e e^2/m\epsilon_0$, e and m are the charge and mass of the electron, and n_e is the electron density. The rapidly decreasing index due to the creation of a plasma leads to a blueshifting and broadening of the spectrum [19]. However, this effect alone cannot change the temporal envelope of the pulse. Self-steepening will also occur due to the rapidly changing plasma index of refraction. The sign of this self-steepening term is the same as self-steepening in a normal material, and therefore it also leads to a steep trailing edge on the pulse but should not decrease pulse duration. Table I summarizes the potential 1D pulse-shaping mechanisms and the characteristic propagation length over which these effects becomes significant [20]. Plasma effects were calculated assuming the total change of the index due to the double ionization of all of the argon. From Table I, it is clear that the plasma-induced phase modulation and self-steepening will be dominant, with some contribution due to conventional SPM. However, no self-compression is possible due to any of the effects listed in Table I.

Since 1D nonlinear wave propagation cannot explain this phenomenon, and since experimentally we found the waveguide to be essential, a 3D model was used to explain the results. Propagation of the laser in the waveguide was modeled by a numerical solution of a 3D scalar wave equation for the laser field, as reported previously [21]. The model takes into account power dissipation due to ionization, and dispersion due to the transient plasma, neutral gas, and the waveguide geometry. Additionally, waveguide loss was measured experimentally and incorporated into the model.

The theoretically predicted pulse durations as a function of pressure are shown in Fig. 3(a), together with the experimentally measured values for comparison [Fig. 3(b)]. The model is in excellent agreement with

TABLE I. Table of possible pulse-shaping mechanisms that involve only 1D propagation effects in Ar at a pressure of 4 torr (i.e., not spatiotemporal pulse evolution effects) and the length of propagation over which they become significant [20]. The calculated gas terms are lower limits calculated for neutral Ar. Key: T , pulse duration; GVD, group velocity dispersion; n_2 , nonlinear index of Ar; n , linear index of Ar; I , laser intensity; ω , laser frequency; c , speed of light; u_{nm} , waveguide mode index; a , waveguide radius.

Mechanism	Length scale	Distance
Waveguide GVD	$\frac{T^2 a^2 \omega^3}{8 \log(2) \mu_{nm}^2 c}$	1 m
Gas GVD	$\frac{T^2}{4} \frac{2\pi c^2}{\log(2) \lambda^3 d^2 n/d\lambda^2}$	1.5 km
Plasma GVD	$\frac{T^2 c \omega}{4 \log(2)} \frac{\omega_p^2}{\omega_p^2}$	60 cm
Gas SPM	$\frac{c}{\omega n_2 I}$	10 cm
Gas self-steepening	$\frac{c T}{n_2^{(3)} I}$	3 m
Plasma blue-shift	$\frac{2 \omega c}{\omega_p^2}$	2.3 mm
Plasma self-steepening	$2 T c \frac{\omega_p^2}{\omega_p^2}$	6.5 cm

experimental data, predicting that the pulse undergoes temporal reshaping that reduces the pulse width from 30 to approximately 13 fs with increasing pressure, and that the minimum pulse duration is obtained around 4 torr. The leading edge of the pulse also steepens in time, accompanied by a longer trailing edge that develops a shoulder as the pressure is increased—again in very good agreement with experiment. The model also predicts less compression for larger diameter waveguides and little or none for the case of no waveguide, as is the case experimentally. Finally, the corresponding pulse spectra blue-shift and increase in bandwidth due to the combined effects of rapidly changing plasma index and self-phase modulation, also in good agreement with experiment. At high pressures above ≈ 8 torr, the pulse splits and the spectrum develops a large long-wavelength wing, leading to the breakup also observed in experiment. The only parameter in our calculations that was adjusted to obtain agreement is the input diameter of the focused laser mode into the waveguide.

From our models, we can identify the mechanism behind the self-compression as a spatiotemporal interplay of plasma-induced refraction and blueshifting, diffraction of the laser beam, phase modulation due to the rapidly changing plasma index, and finally guiding by the hollow waveguide. The role of plasma defocusing [22] is very important in the compression process because it is responsible for the spatiotemporal coupling at high (i.e., ionizing) laser intensities. The presence of the waveguide is also crucial, both experimentally and theoretically. Our models show that as the pulse begins to ionize the gas the trailing edge of the pulse refracts from the center of the waveguide due to plasma-induced defocusing. However, this portion of the beam is then reflected back into the guide by reflection from the waveguide walls. The longer path length traveled will lead to a long trailing edge on

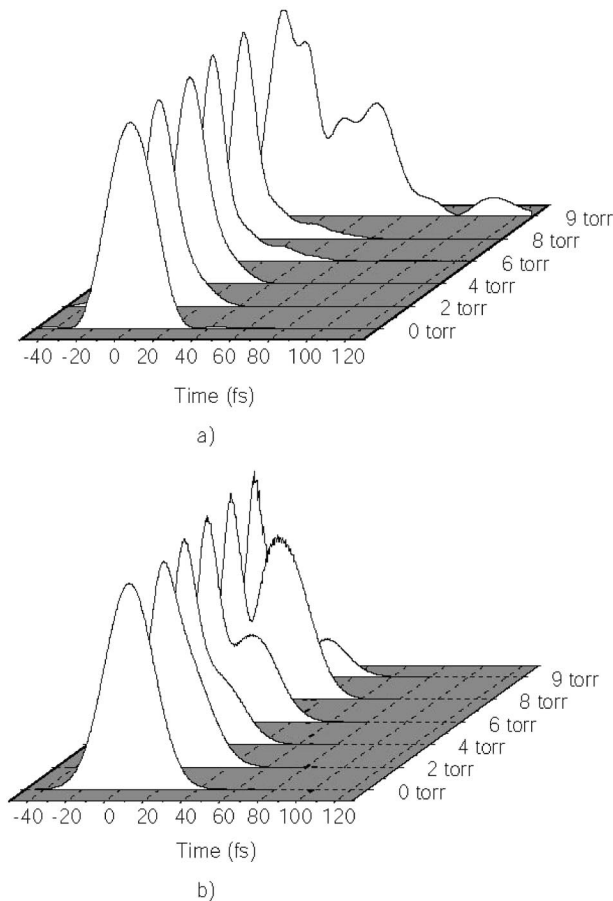


FIG. 3. Comparison between the experimentally observed and theoretically predicted pulse envelopes as a function of pressure for 0, 2, 4, 6, 8, and 9 torr. (a) Experimentally measured pulse shapes. (b) Theoretically predicted pulse shapes. The pulse compression and pulse splitting behavior are seen both experimentally and theoretically, with excellent qualitative and good quantitative agreement between experiment and model.

the pulse. Ionization-induced depletion could explain the sharper leading edge on the pulse, but other effects may also contribute and will be studied. These combined effects lead to a temporally compressed pulse with a sharp leading edge and flat temporal phase, in dramatic contrast to other pulse compression schemes. The reshaping also leads to a small periodic oscillation in the beam diameter and pulse width as the pulse propagates down the waveguide. Both experimentally and theoretically, the pulse compression process is sensitive to the initial mode launched into the waveguide. The laser beam must be focused to a tighter focal spot ($50\text{ }\mu\text{m}$ FWHM) than ideal ($\approx 60\text{ }\mu\text{m}$ FWHM) for coupling into the lowest-order mode of $150\text{ }\mu\text{m}$ waveguide in order to launch a mode that gives rise to large ionization levels and a high degree of plasma-induced refraction.

In conclusion, we demonstrate a new and unanticipated mechanism for pulse compression that operates at laser

intensities above the ionization threshold for the first time. By propagating intense, femtosecond pulses inside a hollow-core waveguide filled with low-pressure argon gas, we demonstrate an increased bandwidth as well as temporal self-compression of a pulse without the need for dispersion compensation. In contrast to previous work, our experiment shows that it is possible to *take advantage of* 3D pulse evolution to compress a light pulse in a way that is not predicted in 1D simulations. Thus, our work provides impetus for further development of 3D modeling of laser-plasma interactions, demonstrating that fundamentally new physical phenomena can be found. This result represents a significant simplification over other pulse compression techniques at laser intensities above the ionization thresholds of atoms and molecules, and will be useful in many applications in high field and plasma science. This mechanism is likely to scale to higher intensities and pulse energies, and likely also to the use of plasma-based waveguides.

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