Self-Compression of High-Intensity Femtosecond Optical Pulses and Spatiotemporal Soliton Generation

I. G. Koprinkov,* Akira Suda, Pengqian Wang, and Katsumi Midorikawa

RIKEN (The Institute of Physical and Chemical Research), Hirosawa 2-1, Wako-shi, Saitama 351-0198, Japan

(Received 17 August 1999; revised manuscript received 20 January 2000)

Self-compression of high-intensity femtosecond pulses has been observed in a number of atomic and molecular gases and solid bulk material. The evolution of the femtosecond pulse parameters during the self-compression has been studied under a variety of experimental conditions. Generation of spatiotemporal solitons has been achieved by the combined action of self-compression and self-focusing.

PACS numbers: 42.65.Tg, 42.65.Re, 42.65.Sf

The propagation of high-intensity femtosecond optical pulses in a nonlinear medium is a complex $(3 + 1)$D phenomenon leading to substantial space-time pulse rearrangement by a number of self-action processes. This problem has attracted considerable attention with the generation of high-intensity femtosecond pulses and a large number of numerical simulations have been performed [1–6]. They generally consider the nonlinear Schrödinger equation (NLSE) with nonlinearity truncated after the cubic term. These works, considering mainly the normal group-velocity dispersion (GVD) $\beta_2 \equiv \partial^2\beta/\partial\omega^2 > 0$, predict a complicated behavior such as self-steeping and splitting (instead of continuous broadening) of the femtosecond pulses when the spatial effects such as diffraction and self-focusing (SF) are also taken into account. The inclusion of higher order effects, e.g., third order dispersion $\partial^3\beta/\partial\omega^3$, "space-time focusing," etc., leads to multiple splitting and coalescence of the split pulses [5]. These predictions have been generally confirmed by experiments on solid bulk materials (BK-7 glass and fused silica [4,5]). Ionization has been also considered as a mechanism influencing femtosecond pulse propagation [6]. In spite of the intensive theoretical studies, the space-time behavior of high-intensity femtosecond pulse propagation is not yet completely understood because, first, the influence of nonlinearities higher than the third order is not well studied in theory and, second, the experiments in that field, which may support the numerical simulations, are insufficient and do not cover a wide range of experimental conditions.

On the other hand, the propagation of the short laser pulses along a 1D medium is a well studied phenomenon, which, in the case of anomalous GVD $\beta_2 < 0$, can lead to temporal soliton formation [7,8]. At normal GVD, no (bright) soliton formation can be achieved based only on $\chi^{(3)}$ nonlinearity, but instead, pulse broadening, intensity shock, and wave breaking could be observed [9]. Considering cubic-quintic NLSE for a 1D medium with $\chi^{(3)} > 0$, $\chi^{(5)} < 0$, a solitary wave solution has been found in positive GVD regime [10]. Such $\chi^{(3)}$, $\chi^{(5)}$ are feasible at given conditions [11]. The existence of spatiotemporal solitons (STS) has been predicted at negative GVD regime [1]. Recently, a STS in one transversal dimension and time, called a $(1 + 1 + 1)$D soliton, has been observed for the first time in $LiIO_3$ by cascading $\chi^{(2)}$ processes producing an effective $\chi^{(3)}$ process [12].

Here we report about a unique behavior of the high-intensity femtosecond pulse—self-compression (SC) of the femtosecond pulse at normal GVD conditions. We have observed for the first time the SC in different optical media—a number of atomic (Ar, Kr) and molecular (CH_4) gases and solid bulk material (fused silica). This reveals the SC as a general phenomenon, which can be considered as the missing temporal counterpart of the SF. From the other side, the SC represents a new compression method scalable for high-energy femtosecond pulses, which does not require anomalous GVD conditions and/or subsequent compression stage. About 10 times pulse compression has been achieved. The SC results in the formation of full-dimensional STS, the so-called "light bullet" [1]. To the best of our knowledge, this is the first experimental generation of the light bullet.

During the course of the present work, we learned about another study [13] reporting twofold SC of high-intensity femtosecond pulse propagation in air. While this could be the same phenomenon, it was only briefly mentioned and no details were given.

The experiments were carried out using a femtosecond Ti:S oscillator-amplifier laser system tuned at 750 nm. The output from the oscillator (Spectra-Physics, Tsunami) was amplified by a regenerative amplifier (BMI, α-10), operating at a 10-Hz repetition rate. The pulses after the regenerative amplifier were compressed by a grating compressor to 150 fs and had typically 1-mJ pulse energy. To ensure control of the medium parameters, we used pressurized static gases in an 80-cm-long cell. The intensity at the focal point was estimated at 2.5×10^{13} W/cm^2, assuming no SF. The pulse duration was measured from the autocorrelation traces using a noncollinear single-shot second harmonic autocorrelation scheme with a 100-μm-thick beta barium borate crystal and monitored by a diode array. The spectrum was analyzed using a charge-coupled device equipped grating spectrometer with 0.8-nm resolution.

We carried out a number of experiments concerning the spatiotemporal and spectral behavior of the high-intensity femtosecond pulse propagation in different media. The main features were found to be similar for all of the media studied. The experimental values of the pulse duration, spectral bandwidth, and the corresponding transform-limited pulse duration calculated for sech^2 pulse are shown in Fig. 1 as a function of gas pressure. At zero pressure, the extension of the pulse duration above 150 fs results from the transmittive optics. With increasing gas pressure, the pulse duration initially increases due to the normal GVD conditions, but just above 2 atm of CH_4, Fig. 1(a), a rapid collapse is observed, thus shortening the pulse duration to 40–50 fs. The beginning of the temporal collapse can be clearly related to the spatial collapse due to SF. The shortest pulse due to the SC was about 30 fs. This gives about 10 times pulse compression relative to the pulse duration just before the onset of the SC. In the same time, the spectral bandwidth, measured around the half maximum, broadens not so much due to self-phase modulation (SPM)—from initial bandwidth of (6.5–7) THz to (9–10) THz. Strong spectral rearrangement and generation of well-structured white-light supercontinuum of more than 170 THz bandwidth (FWHM) have also been observed. However, its intensity (pulse energy ≈ 1 μJ) was negligible in comparison with the intensity of the main spectrum. That is why only the near frequency structure of the main spectrum is involved in the SC. The pulse reaches the transform-limited pulse duration for sech^2 pulse at 6 atm. Further shortening was restricted by the spectral bandwidth.

Figure 1(b) shows the SC in Ar, having $\beta_2 \approx +0.6$ fs^2/cm positive GVD at about 4 atm [14]. While the trend is almost the same as in CH_4, the lower nonlinearity of Ar makes the process much more gradual with pressure. A very broad pressure range (6–14 atm) exists in Ar without shoulders or splitting of the optical pulse. Pulse splitting in Ar occurs at about 18 atm. In Kr ($\beta_2 \approx +0.6$ fs^2/cm at 2 atm [14]), once the SC takes place, the pulse duration remains nearly constant between 4 and 10 atm. No splitting has been observed up to 15 atm while a strong pedestal appears at this pressure. Some preliminary results concerning SC in Ar and Kr will appear in Ref. [15].

To ensure a wide pressure range for changing the material parameters, the above experiments were performed using 10-mm-thick cell windows. To diminish the effect of the cell windows, experiments using 1-mm-thick windows were also done. The behavior generally remains the same, Fig. 2, but no initial pulse broadening was observed due to the reduced positive chirp introduced by the thin windows. Figure 2 also illustrates the SC at different pulse energies, 0.7 and 1 mJ. Both dependencies appear similar, but increasing the pulse energy leads to a systematic shift of the process toward lower pressures. Thus, the SC of the pulse can be triggered by increasing either the medium nonlinearity $\chi^{(n)}$ [gas pressure (Fig. 1)] or the field intensity [energy (Fig. 2)]. These determine the magnitude of the nonlinear field-matter interaction.

Finally, the SC of the femtosecond pulses has also been observed in fused silica. In contrast to other experiments on solid material where a small portion of the beam is focused on the entrance surface, we used the full pulse energy (1 mJ) while keeping the sample far beyond the focal point. At $\beta_2 \approx +470$ fs^2/cm, including material ($\approx +400$ fs^2/cm) and virtual waveguide ($\approx +70$ fs^2/cm) contributions at 750 nm, we observed twofold pulse SC when the sample approached the focal point from 50 to 15 cm. The observed SC (instead of splitting [4,5]) is attributed to the higher intensity used in our experiment.

The above experiments demonstrate that the SC of the high-intensity femtosecond pulses is a *general phenomenon* that can be observed in a variety of optical media under suitable excitation conditions.

The SC was accompanied by a marked improvement of the pulse shape. The evolution of the pulse shape (autocorrelation traces) with CH_4 pressure is shown in Fig. 3. It closely resembles the formation of a temporal soliton in optical fibers [8]. However, instead of a real fiber, we have an optically written virtual fiber in the bulk medium due to the SF. From 3 to 5 atm the pulse shape stays the same—nearly the sech^2 shape of the fundamental soliton. Increasing the gas pressure leads to splitting preceded by the pedestal formation. At normal GVD, the SC is the

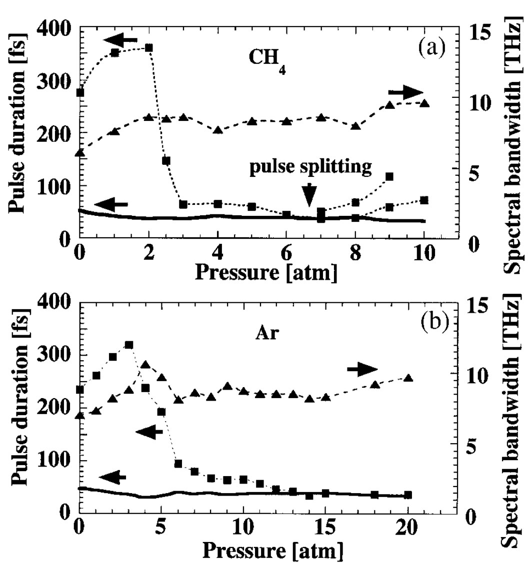

FIG. 1. Pulse duration, spectral bandwidth, and the corresponding transform-limited pulse duration (full line) versus pressure in CH_4 (a) and Ar (b).

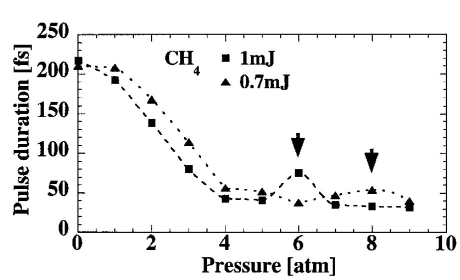

FIG. 2. Pulse duration versus CH_4 pressure at different pulse energies. The increasing of the pulse duration marked by the arrow results from formation of pedestal.

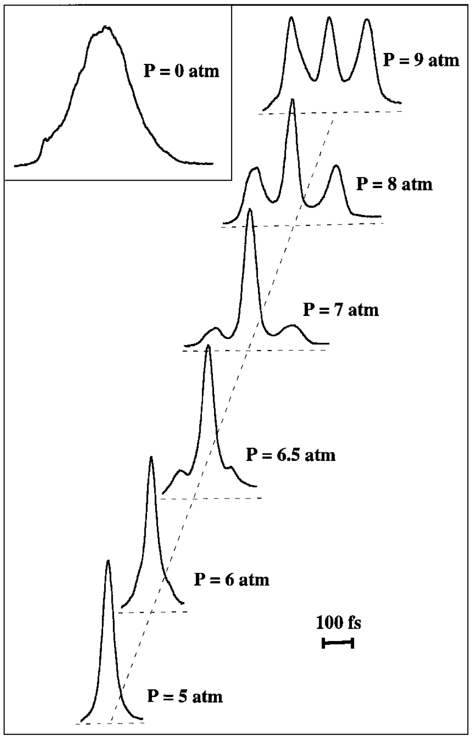

FIG. 3. Evolution of the pulse shape (autocorrelation traces) with CH_4 pressure. The inset shows the autocorrelation trace of the laser pulse passing through the empty cell, $P = 0$ atm.

confinement mechanism forcing the pulse toward the temporal soliton formation—soliton compression.

Because of the SF, a light filament was formed, trapping about 40% of the pulse energy. The formation of stable light filament is an evidence for spatial soliton formation. The spatial collapse of the beam, followed by a substantial improvement of the beam quality, is shown in Fig. 4. The beam intensity profile keeps stable between 3 and 6 atm of CH_4 pressure. Some amount of light trapping in CH_4 persists up to 40 atm.

The study of the spatiotemporal pulse stability is complicated due to the inherent limitations in a real experiment. In particular, the higher order cubic-quintic solitary waves were generated ($\beta_2 < 0$) and tested for stability only numerically [16]. As mentioned above, the spatial and temporal intensity distributions were found stable within a given pressure range. The "robustness" of the optical pulse can be deduced from its pressure behavior because changing the pressure to a certain degree simulates the propagation effect. The nearly constant pulse duration of stable shape after the SC takes place, Fig. 1, can be interpreted in such a sense. This is better expressed in the case of Ar. The pulse was also stable along the light filament. The latter was verified by moving the cell along the beam, thus changing the length of the light filament. The pulse duration varies insignificantly with the position of the cell, Fig. 5, provided the focal point is far from the entrance window so as to avoid strong SPM in the window material.

Based on the above results, we may conclude that full space-time self-trapping of the light matter is achieved having a stable self-sustained behavior within a given range of material parameters. This will be called full-dimensional STS. The SC and SF are the responsible phenomena for the STS formation in the temporal and spatial aspects, respectively. It is noteworthy that the observed spatiotemporal collapse appears as a natural feature of light that does not require sophisticated external constraint. In contrast to [12], we achieved complete space-time confinement of the optical pulse. In addition, this was realized in suitable for high-intensity STS generation isotropic media, avoiding the problems with the irreversible medium damage, phase-matching, and negative GVD.

From a phenomenological point of view, the SC resembles the SF and can be thought of as being a time-domain counterpart of the SF. However, some substantial differences between these phenomena exist. The SF results from positive $\chi^{(3)}$ to compensate for the diffraction and, eventually, for high-intensity fields, the self-defocusing due to higher order negative $\chi^{(n)}$ ($n > 3$) [11,17], and/or ionization induced refractive index change.

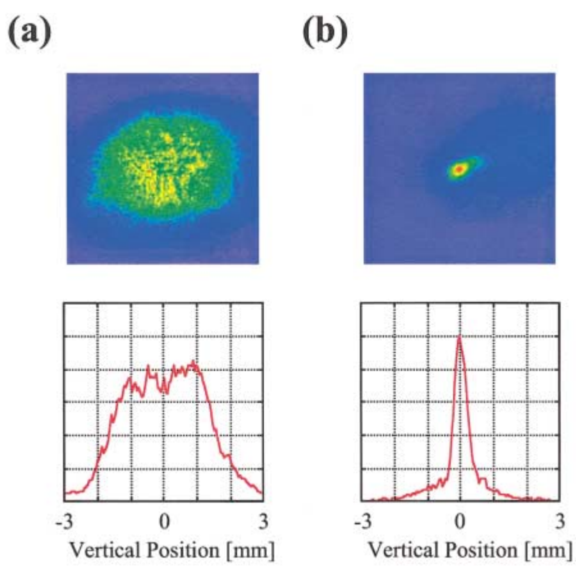

FIG. 4 (color). Beam spots and the corresponding transversal intensity profiles in CH_4 at $P = 0$ atm (a) and $P = 3$ atm (b) after the SF and SC occur, taken 30 cm from the exit cell window.

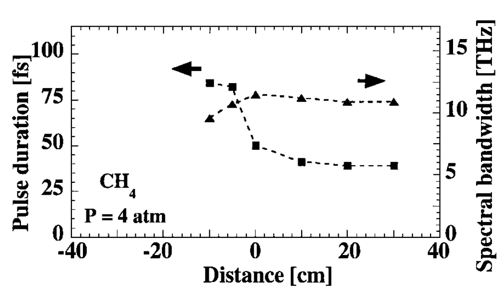

FIG. 5. Pulse duration versus distance between position of the focal point (when the cell is removed) and the position of the cell center.

In the case of SC we have a quite different situation. As the numerical simulations show [2–5], the combination of positive $\chi^{(3)}$ and positive GVD does not lead to pulse shortening without splitting. In our case, the shortening appears as a primary process that does not result from splitting but from the increased nonlinear field-matter interaction. Thus, the observed SC should be distinguished from $\chi^{(3)}$ dominated nonlinear processes. However, a suitable value of the higher order susceptibilities (e.g., $\chi^{(5)} < 0$) may arrest the pulse spreading/splitting leading to solitary wave formation even at positive GVD [10]. This results from the fast dynamic interplay between dispersion and intensity dependent factors, i.e., the nonlinear refractive indexes $n_2|E|^2$ ($n_2 = 3\chi^{(3)}/8n_0$) and $n_4|E|^4$ ($n_4 = 5\chi^{(5)}/16n_0$), which appear in the respective terms of the NLSE. Because of the quadratic intensity dependence, the second term may become comparable to or even exceed the first one for high-intensity fields. The SF, as it leads to an increase of the pulse intensity, simply triggers the SC. This is the origin of the close relation between the SC and the SF. The role of the intensity in the self-trapping of the optical pulse can be qualitatively illustrated with Fig. 6 showing multiple splitting in Ar at 20 atm. As can be seen, one splits the longer, but low-intensity pulse instead of the shorter, but high-intensity one, which should be the case, based only on simple dispersion considerations. The factors leading to the STS and SC at positive GVD must be the same, but in the first case they are well balanced while in the second, the compression factor dominates. We consider that the above is the basic mechanism of the observed SC and STS. Such a mechanism, including only fast electronic processes, is universally applicable to all media. Although suitable $\chi^{(5)}$ appears to be sufficient to explain the SC and the STS, actually they result from the entire dynamic contribution of all orders of self-action processes. This leads to self-consistent nonlinear field-matter interaction, whose optical counterpart is the STS. Negative change of the refraction index results also from ionization. Despite the fact that free electron density does not instantaneously follow the field intensity, it may also cause fast effects on the field by means of secondary processes such as dispersion and defocusing.

The different outcome in the "low-intensity" ($\cong 10^{11}$ W/cm^2—Refs. [4,5]) and "high-intensity" ($>10^{13}$ W/cm^2—our case; and $>10^{14}$ W/cm^2—Ref. [13]) experiments shows that the behavior of the femtosecond pulse strongly depends on its intensity (more precisely, the magnitude of the nonlinear field-matter interaction). Thus, the problem of high-intensity femtosecond pulse propagation requires including in the (3 + 1)D NLSE not only higher order dispersion [5] and ionization [6,13], but also higher order nonlinearity [10].

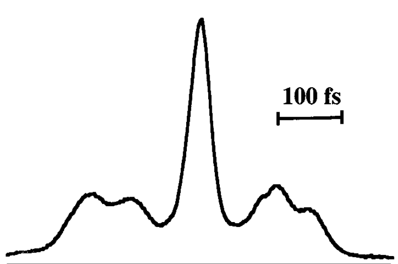

FIG. 6. Multiple pulse splitting in Ar at 20 atm.

In conclusion, self-compression of high-intensity femtosecond pulses has been found for the first time in a variety of optical media. This reveals the self-compression as a general phenomenon, which can be considered as the temporal counterpart of the self-focusing. A full-dimensional spatiotemporal soliton has been generated for the first time as a result of the combined action of the self-compression and the self-focusing.

I. G. K. gratefully acknowledges the hospitality and the financial support from the Laser Tech. Lab–RIKEN in the performance of the experiments.

*Permanent address: Institute of Applied Physics, Technical University of Sofia, 1756 Sofia, Bulgaria.
Email address: igk@vmei.acad.bg

[1] Y. Silberberg, Opt. Lett. **15**, 1282 (1990).
[2] P. Chernev and V. Petrov, Opt. Lett. **17**, 172 (1992).
[3] J. E. Rothenberg, Opt. Lett. **17**, 583 (1992); **17**, 1340 (1992).
[4] J. K. Ranka, R. W. Schirmer, and A. L. Gaeta, Phys. Rev. Lett. **77**, 3783 (1996); J. K. Ranka and A. L. Gaeta, Opt. Lett. **23**, 534 (1998).
[5] S. A. Diddams, H. E. Eaton, A. A. Zozulya, and T. S. Clement, Opt. Lett. **23**, 379 (1998); A. A. Zozulya, S. A. Diddams, A. G. Van Engen, and T. S. Clement, Phys. Rev. Lett. **82**, 1430 (1999).
[6] O. G. Kosareva, V. P. Candilarov, A. Brodeur, C. Y. Chien, and S. L. Chin, Opt. Lett. **22**, 1332 (1997).
[7] A. Hasegawa and F. Tappert, Appl. Phys. Lett. **23**, 142 (1973).
[8] F. L. Mollenauer, R. H. Stolen, and J. P. Gordon, Phys. Rev. Lett. **45**, 1095 (1980).
[9] J. E. Rothenberg and D. Grischkowsky, Phys. Rev. Lett. **62**, 531 (1989).
[10] S. Tanev and D. Pushkarov, Opt. Commun. **141**, 322 (1997).
[11] H. Puel, K. Spanner, W. Falkenstein, W. Kaiser, and C. R. Vidal, Phys. Rev. A **14**, 2240 (1976).
[12] X. Liu, L. J. Quin, and F. W. Wise, Phys. Rev. Lett. **82**, 4631 (1999).
[13] H. R. Lange *et al.*, in *Proceedings of the XIth International Conference on Ultrafast Phenomena, 1998* (Springer, Berlin, 1998), p. 115.
[14] M. Nisoli, S. De Silvestri, and O. Svelto, Appl. Phys. Lett. **68**, 2793 (1996).
[15] I. G. Koprinkov, Akira Suda, Pengqian Wang, and Katsumi Midorikawa, Jpn. J. Appl. Phys. (to be published).
[16] S. Cowan, R. H. Enns, S. S. Rangnekar, and S. S. Sanghera, Can. J. Phys. **64**, 311 (1986).
[17] S. A. Akhmanov, A. P. Sukhorukov, and R. V. Khokhlov, Soviet Phys. JETP **23**, 1025 (1966).