# Observation of Pulse Splitting in Nonlinear Dispersive Media

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We investigate experimentally the nonlinear dynamics of a femtosecond pulse undergoing self-focusing in a normally dispersive medium. Above a certain threshold power, we observe that the pulse splits into two pulses, and measurements of the pulse spectra provide evidence that supercontinuum generation is closely connected with the temporal splitting. We also observe that the split pulses can undergo additional splittings. Our observations are in good agreement with the predictions of the theoretical model based on the nonlinear Schrödinger equation describing self-focusing in dispersive media. [S0031-9007(96)01571-2]

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The self-focusing of a light beam is one of the most fundamental nonlinear optical processes. In a material with an index of refraction that increases with intensity (i.e., $n = n_0 + n_2 I$, where $n_2 > 0$), a steady-state analysis predicts that a laser beam will self-focus if the power $P$ in the beam is greater than the critical power $P_{\mathrm{cr}} = \pi(0.61\lambda)^2/8n_0n_2$ [1]. The steady-state paraxial wave-equation model predicts that for a sufficiently long interaction length and $P > P_{\mathrm{cr}}$, a singularity results which corresponds to catastrophic self-focusing. Under these conditions, several optical processes (other than optical damage to the material) can occur, which can lead to the arrest of catastrophic self-focusing, to filament formation [2], or to the production of multiple foci [3]. These processes may include avalanche breakdown [4], saturation of the nonlinear index [5], nonlinear absorption [6], or diffraction in the nonparaxial limit [7,8]. In the absence of material dispersion, the steady-state model of self-focusing can be extended for pulses through the use of the moving-focus model [9]. In addition, it was shown [10] that self-focusing can lead to temporal shortening of the transmitted pulse when the length of the medium is nearly equal to the self-focusing length.

For pulses in the femtosecond regime, the effects of material dispersion might be expected to play an important role in self-focusing. Recent theoretical studies [11–18] demonstrate that the inclusion of group-velocity dispersion (GVD) dramatically modifies the dynamics of the process. Using the nonlinear Schrödinger equation (NLSE) in the positive GVD regime, it is found that above a certain threshold power the pulse undergoes temporal splitting which can arrest catastrophic self-focusing. At higher powers, it is still an open question as to whether these split pulses can undergo additional splittings [14,17–19]. Remarkably, the pulse splittings are predicted to occur even when dispersion might be thought to be negligible as compared to the nonlinearity. Additional analyses [15,20] in the negative GVD regime have been undertaken, and the possibility of the formation of three-dimensional solitons has been considered [20].

In parallel with the research on self-focusing, extensive studies have shown that the interaction of intense, ultrashort light pulses with matter can result in the generation of optical radiation over an extremely broad spectral range [21]. This phenomenon, known as supercontinuum generation (SCG) or white-light generation, was first observed [22] in 1970, and since then it has been observed in many different solids [23], liquids [24], and gases [25,26] under a wide variety of experimental conditions. The shape of the spectra generated in various media are strikingly similar, which suggests that SCG is a universal feature of laser-matter interactions. Nevertheless, the underlying mechanisms of SCG are still not well understood. Corkum, Rolland, and Srinivasan-Rao [25] first noted the connection between self-focusing and SCG, and, more recently, it was suggested [27] that SCG may occur as a result of the interplay between self-focusing, self-phase modulation, and GVD. Although numerical studies [11,13] using the NLSE appear to support this hypothesis, there has not been any experimental verification of the main predictions of this model.

In this Letter, we describe the results of an experimental study of self-focusing in a normally dispersive medium. In agreement with the theoretical predictions of the NLSE, we observe that an ultrashort pulse undergoes pulse splitting for input powers that are above the critical power. Above the threshold power for pulse splitting, the pulse spectrum undergoes significant broadening which eventually develops into SCG at higher powers. We also find that, under suitable conditions, the split pulses can themselves undergo further splitting. We have concentrated our studies over a range of input powers that are near the threshold for temporal splitting, since, in this regime, comparisons with numerical simulations of the NLSE can be made.

The proposed theoretical model for describing self-focusing of femtosecond pulses in a dispersive medium is based on the following NLSE for the slowly varying amplitude $A(\mathbf{r}, t)$ of an optical pulse centered at frequency $\omega$:

$$\frac{\partial A}{\partial z} - \frac{i}{2k} \nabla_\perp^2 A + i \frac{\beta_2}{2} \frac{\partial^2 A}{\partial \tau^2} = i \frac{n_2 n_0 \omega}{2\pi} |A|^2 A , \quad (1)$$

where $\beta_2$ is the GVD, $k = n_0\omega/c$ is the wave-vector amplitude, and $\tau = t - z/v_g$ is the retarded time for the pulse traveling at the group velocity $v_g$. This equation has been studied primarily for the case in which the beam has radial symmetry (i.e., $\nabla_\perp^2 = \partial^2/\partial r^2 + \partial/r\partial r$). For the case in which the input pulse is Gaussian in space and time such that $A(r,t) = A_0 \exp[-r^2/2w_0^2 - t^2/2\tau_p^2]$, three length scales can be used to characterize the interaction, the diffraction length $L_{\rm DF} = kw_0^2/2$, the dispersion length $L_{\rm DS} = \tau_p^2/|\beta_2|$, and the nonlinear length $L_{\rm NL} = (n_2 n_0 \omega |A_0|^2/2\pi)^{-1}$. The ratio $L_{\rm DF}/L_{\rm NL}$ can be related to the ratio of the input peak power $P$ to $P_{\rm cr}$ through the expression $P/P_{\rm cr} = 1.885 L_{\rm DF}/L_{\rm NL}$ [12]. We numerically integrate Eq. (1) using standard techniques [13], and in Fig. 1 the predicted on-axis (i.e., at $r = 0$) temporal profile, frequency chirp, and spectrum of the transmitted pulse are shown for increasing input powers. For these simulations the length $L$ of the medium is $L = 1.5 L_{\rm DF}$ and $L_{\rm DF}/L_{\rm DS} = 0.04$. At low powers ($P/P_{\rm cr} = 0.5$), the temporal profile [Fig. 1(a)] is nearly identical to the input pulse since $L \ll L_{\rm DS}$. The shape of the frequency chirp is similar to what would be expected from pure self-phase modulation. The corresponding spectrum [Fig. 1(a')] is slightly broader than that of the input pulse. Figure 1(b) shows the temporal profile at the onset ($P/P_{\rm cr} = 1.4$) of pulse splitting. It is apparent that the chirp is highly nonlinear and that the center frequency of the leading (trailing) pulse is downshifted (upshifted) from the initial frequency of the incident pulse [12,13]. The broadening of the on-axis spectrum [Fig. 1(b')] is apparent, and the spectrum begins to develop oscillatory structure with a period of oscillation inversely related to the pulse separation [11]. At higher powers ($P/P_{\rm cr} = 1.5$), the pulses become well separated in time and are considerably shorter than the initial pulse [Fig. 1(c)]. The corresponding spectrum shown in [Fig. 1(c')] shows dramatic broadening with clear evidence of oscillations. As the power is increased further, the split pulses become shorter in time, which indicates that the inclusion of higher-order dispersion terms is necessary and may also signal a breakdown of the slowly varying envelope approximation. It has been proposed that the extremely short duration of these pulses is responsible for SCG [11,13]. As a result of the enormous computing resources required by these types of simulations, we are not able to determine whether or not the split pulses will continue to self-focus and undergo further splittings.

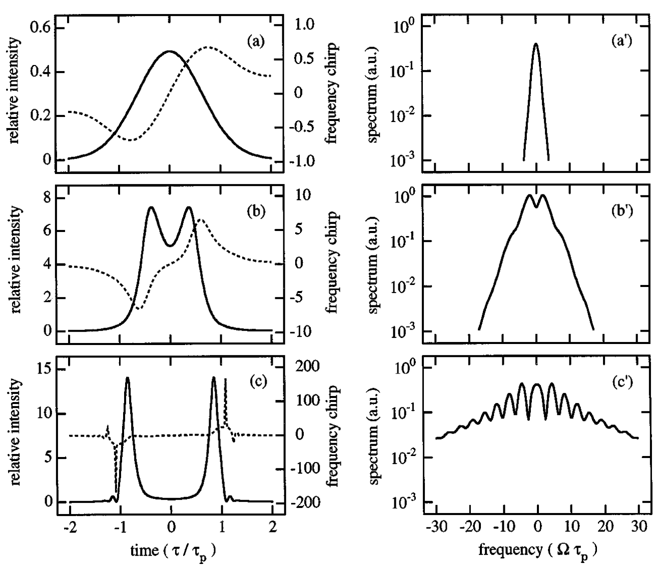


FIG. 1. Theoretical predictions for the on-axis temporal profile (solid line) and chirp (dashed line) (a)–(c) and for the spectrum (a')–(c') of the transmitted beam as the input power is increased. The ratio of the peak power of the incident pulse to the critical power in each pair of plots is (a),(a') 0.5; (b),(b') 1.4; and (c),(c') 1.5.

We have carried out a series of experiments to test the predictions of the nonlinear Schrödinger equation model. The pulses were produced by a Ti:sapphire regenerative laser system (Clark-MXR) operating at 800 nm. This system generated 1-mJ pulses at a 1-kHz repetition rate and with a duration in the range 85–90 fs. The shot-to-shot fluctuations of the pulse energy were less than 6%. The beam was apertured slightly which resulted in a near-Gaussian transverse profile. A small fraction of the output was focused to a 55 $\mu$m spot size near the front face of a BK7 glass window, which served as the nonlinear dispersive medium. The value of the GVD parameter for BK7 is $\beta_2 = 446$ fs$^2$/cm, and the value of the nonlinear index coefficient [28] is $n_2 = 3.45 \times 10^{-16}$ cm$^2$/W which corresponds to $P_{\rm cr} = 1.8$ MW. In the studies presented here, we used both 0.5- and 1-in.-thick samples. Using a series of lenses and apertures, we analyzed the temporal and spectral behavior of the central part of the transmitted beam as the power was increased above the threshold power for temporal splitting. The temporal autocorrelation of the pulses was taken using a scanning ($\sim$1-sec-long scan) autocorrelator in a noncollinear geometry with a 300-$\mu$m-thick BBO crystal. Our system is capable of accurately measuring transform-limited input pulses as short as 25 fs. The spectrum was acquired concurrently using a fiber-coupled 0.275-m spectrometer with 0.5-nm resolution.

Figure 2 shows the autocorrelation and the corresponding spectrum of the transmitted pulses as the incident power was increased from well below to above the threshold power for pulse splitting. These data were taken with the 1-in. glass sample, and the input-pulse duration was 85 fs. At relatively low powers ($P = 0.6$ MW,

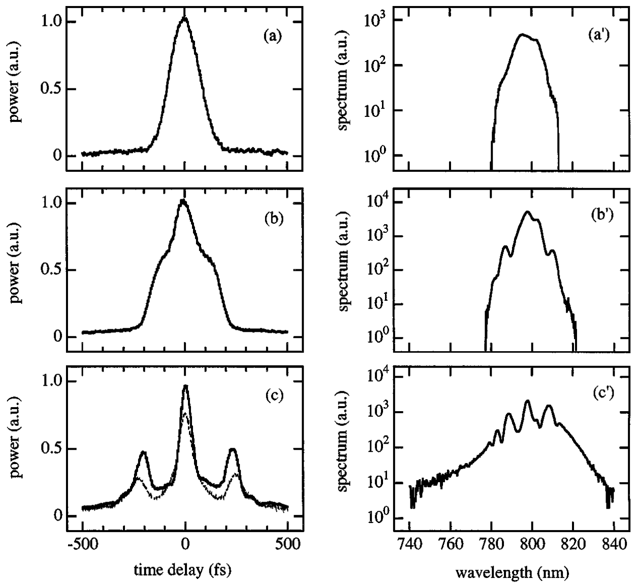


FIG. 2. Experimentally measured temporal autocorrelations (a)–(c) and power spectra (a')–(c') of the pulses transmitted through a 1-in. BK7 glass sample as the input power is increased. The peak power for each set of plots is (a),(a') 0.6 MW; (b),(b') 3 MW; and (c),(c') 4.8 MW. The dotted curve in (c) is the autocorrelation of the transmitted beam after it is passed (unfocused) through an additional piece of BK7 glass. From the increase in the pulse separation, we infer that the central wavelengths of the split pulses differ by 16 nm.

$P/P_{\rm cr} = 0.33$), the temporal profile [Fig. 2(a)] and the spectrum [Fig. 2(a′)] of the transmitted pulse are nearly identical to those of the incident pulse. The appearance of shoulders on the autocorrelation shown in Fig. 2(b) is the signature of the threshold ($P = 3$ MW, $P/P_{\rm cr} = 1.7$) for pulse splitting. The observed threshold power for pulse splitting is within 15% of the predicted value for our experimental conditions. The spectrum [Fig. 2(b′)] of the pulse is seen to exhibit broadening and an oscillatory structure. In their studies in high-pressure noble gases, Corkum and Rolland [29] also observed oscillations in the spectrum of the transmitted pulse. At higher powers ($P = 4.8$ MW, $P/P_{\rm cr} = 2.7$), clear evidence of the pulse splitting is observed as illustrated by the three-peaked autocorrelation [solid curve in Fig. 2(c)]. Each of the peaks is considerably narrower than the autocorrelation of the initial pulse, which is consistent with theoretical predictions. We also performed a cross correlation with a split-off portion of the 85 fs beam from the laser system and found that the split pulses in the transmitted field have the same peak power. In order to verify that the pulses are at different central frequencies, we passed the transmitted beam (unfocused) through a 0.5-in. piece of BK7 glass, and measured the temporal autocorrelation of the beam [the dashed curve in Fig. 2(c)]. The temporal separation of the split pulses is seen to increase, as would be expected for pulses having different group velocities.

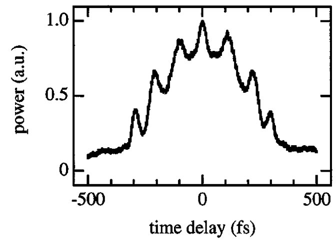


FIG. 3. Experimentally measured temporal autocorrelation of the transmitted beam which shows evidence of secondary splittings. The peak power of the input beam is 7.6 MW.

From this increase in separation, we estimate that the difference between the center wavelengths of the two pulses is 16 nm.

The spectrum [Fig. 2(c′)] of the transmitted beam above the threshold for pulse splitting exhibits substantial broadening and a shape that is characteristic of the spectra of SCG observed in other media [21,23–26], and the generation of white light around the transmitted beam can be seen clearly by eye. At slightly higher powers, the broadening increases explosively. We believe that these observations confirm that supercontinuum generation is a result of the nonlinear dynamics of self-focusing in which the temporal and spatial degrees of freedom are coupled by the presence of GVD. However, treating theoretically the problem of supercontinuum generation far above threshold will require a model in which neither the paraxial assumption nor the slowly varying envelope approximation are made.

We also investigated the issue of whether the split pulses could themselves undergo temporal splitting. With the 1-in. sample, we did not observe clear evidence of additional splittings. However, by using the 0.5-in. sample, we were able to demonstrate that the split pulses can undergo further splittings, as evidenced by the 7-peaked autocorrelation shown in Fig. 3 ($P = 7.6$ MW). In this case, the duration of the incident pulse was 90 fs, and the threshold power for the initial splitting was measured to be 3.3 MW.

These results are relevant for a range of applications such as Kerr-lens mode locking [30], the amplification of femtosecond pulses using regenerative laser amplifiers, and the optical compression of femtosecond pulses [31]. Since supercontinuum generation is used as a source of broadly tunable femtosecond pulses for spectroscopic applications, understanding the mechanisms that lead to SCG may make it possible to tailor the characteristics of the generated light for a particular application. Furthermore, the results shown here demonstrate that small amounts of dispersion may also lead to dramatic changes in the dynamics of intense laser-matter interactions under conditions in which a plasma is generated [32].

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[1] See, for example, *Nonlinear Optics*, edited by R. W. Boyd (Academic, Boston, 1992), Sec. 6.2.
[2] R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Lett. **13**, 479 (1964).
[3] S. A. Akhmanov, R. V. Khokhlov, and A. P. Sukhorukov, in *Laser Handbook*, edited by F. T. Arecchi and E. O. Schulz-Dubois (North-Holland, Amsterdam, 1972); J. H. Marburger, Prog. Quantum Electron. **4**, 35 (1975).
[4] N. Bloembergen, Opt. Commun. **8**, 285 (1973).
[5] J. H. Marburger and E. L. Dawes, Phys. Rev. Lett. **21**, 556 (1968); E. L. Dawes and J. H. Marburger, Phys. Rev. **179**, 862 (1969).
[6] A. L. Dyshko, V. N. Lugovi, and A. M. Prokhorov, Sov. Phys. JETP **34**, 1235 (1972).
[7] M. D. Feit and J. Fleck, Jr., J. Opt. Soc. Am. B **5**, 633 (1988).
[8] G. Fibich, Phys. Rev. Lett. **76**, 4356 (1996).
[9] M. M. T. Loy and Y. R. Shen, Phys. Rev. Lett. **22**, 994 (1969).
[10] J. H. Marburger and W. G. Wagner, IEEE J. Quantum Electron. **QE-3**, 415 (1967); G. L. McAllister, J. H. Marburger, and L. G. DeShazer, Phys. Rev. Lett. **21**, 1648 (1968).
[11] J. E. Rothenberg, Opt. Lett. **17**, 583 (1992).
[12] P. Chernev and V. Petrov, Opt. Lett. **17**, 172 (1992); Opt. Commun. **87**, 28 (1992).
[13] G. G. Luther, A. C. Newell, J. V. Moloney, and E. M. Wright, Opt. Lett. **19**, 789 (1994); G. G. Luther, J. V. Moloney, A. C. Newell, and E. M. Wright, Opt. Lett. **19**, 862 (1994).
[14] G. G. Luther, A. C. Newell, and J. V. Moloney, Physica (Amsterdam) **74D**, 59 (1994).
[15] X. D. Cao, G. P. Agrawal, and C. J. McKinstrie, Phys. Rev. A **49**, 4085 (1994).
[16] A. T. Ryan and G. P. Agrawal, Opt. Lett. **20**, 306 (1995).
[17] G. Fibich, V. M. Malkin, and G. C. Papanicolaou, Phys. Rev. A **52**, 4218 (1995).
[18] L. Bergé, J. J. Rasmussen, E. A. Kuznetsov, E. Shapiro, and S. K. Turitsyn, J. Opt. Soc. Am. B **20**, 1879 (1996).
[19] N. Zharova, A. Litvak, T. Petrova, A. Sergeev, and A. Yunakovsky, Pis'ma Zh. Eksp. Teor. Fiz. **44**, 12 (1986) [JETP Lett. **44**, 13 (1986)].
[20] S. N. Vlasov, L. V. Piskunova, and V. I. Talanov, Zh. Eksp. Teor. Fiz. **44**, 12 (1986) [Sov. Phys. JETP **95**, 13 (1989); Y. Silberberg, Opt. Lett. **15**, 1282 (1990); A. B. Blagoeva, S. G. Dinev, A. A. Dreischih, and A. Naidenov, IEEE J. Quantum Electron. **QE-27**, 2060 (1991).
[21] See, for example, *The Supercontinuum Laser Source*, edited by R. R. Alfano (Springer-Verlag, New York, 1989).
[22] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24**, 584 (1970); **24**, 592 (1970).
[23] P. B. Corkum, P. P. Ho, R. R. Alfano, and J. T. Manassah, Opt. Lett. **10**, 624 (1985); G. Sucha and D. S. Chemla, Opt. Lett. **16**, 1177 (1991).
[24] W. L. Smith, P. Liu, and N. Bloembergen, Phys. Rev. A **15**, 2396 (1977); R. L. Fork, C. V. Shank, C. Hirlmann, and R. Yen, Opt. Lett. **8**, 1 (1983).
[25] P. B. Corkum, C. Rolland, and T. Srinivasan-Rao, Phys. Rev. Lett. **57**, 2268 (1986).
[26] J. H. Glownia, J. Misewich, and P. P. Sorokin, J. Opt. Soc. Am. B **3**, 1573 (1986); T. R. Gosnell, A. J. Taylor, and D. P. Greene, Opt. Lett. **15**, 130 (1990); V. Francois, F. A. Ilkov, and S. L. Chin, Opt. Commun. **99**, 241 (1993).
[27] D. Strickland and P. B. Corkum, Proc. Soc. Photo-Opt. Instrum. Eng. **1413**, 54 (1991).
[28] D. Huang, M. Ulman, L. H. Acioli, H. A. Haus, and J. G. Fujimoto, Opt. Lett. **17**, 511 (1992).
[29] P. B. Corkum and C. Rolland, IEEE J. Quantum Electron. **QE-25**, 2634 (1989).
[30] I. P. Christov, V. D. Stoev, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **20**, 2111 (1995).
[31] M. Nisoli, S. De Silvestri, and O. Svelto, Appl. Phys. Lett. **68**, 2793 (1996).
[32] See, for example, A. Braun, G. Korn, X. Liu, D. Du, J. Squier, and G. Mourou, Opt. Lett. **20**, 73 (1995).