Generation of Optical Spatiotemporal Solitons

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We report the experimental formation of optical spatiotemporal solitons. Intense femtosecond-duration pulses experience a saturable self-focusing nonlinearity due to the cascading of quadratic nonlinear processes. Strong coupling of the interacting fundamental and harmonic waves produces solitons that overcome diffraction in one spatial dimension as well as group-velocity dispersion to reach constant beam size and pulse duration, both compressed with respect to the input values. Spatiotemporal solitons are observed both with nearly zero and with large group-velocity mismatch between fundamental and harmonic pulses. [S0031-9007(99)09288-1]

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Solitons, steadily propagating localized waves, have been studied in many areas of science over the last decade. Solitons that exhibit confinement in one dimension (spatial or temporal) have been the subject of extensive theoretical and experimental investigations [1]. Multidimensional solitons exhibit confinement in more than one transverse dimension. These include stationary waves that are confined in two transverse spatial dimensions as well as spatiotemporal solitons (STS). STS result from the simultaneous balance of diffraction and dispersion by self-focusing and nonlinear phase modulation, respectively. Much theoretical work on multidimensional solitons has been reported [2], but few experimental results exist owing to the difficulty of creating appropriate experimental conditions. Interest in optical STS in solids received much impetus from Silberberg's consideration of their formation in bulk materials of arbitrary dimensions [3]. STS are theoretically unstable in third-order nonlinear media [4], but solutions can be stabilized if the nonlinearity is saturable.

Recently, there has been a resurgence of interest in the effective third-order ($\chi^{(3)}$) nonlinearity that arises from the cascading of second-order ($\chi^{(2)}$) processes. The phase shift arising from back conversion of second-harmonic light under non-phase-matched conditions depends linearly on intensity at low intensities, and so can be modeled as resulting from an effective nonlinear index of refraction $(n_2)_{\rm eff}$. The residual second-harmonic generation (SHG) is analogous to two-photon absorption. The renewed interest is based on the recognition that large effective third-order nonlinearities of controllable sign can be produced in the cascade process [5]. The nonlinearity saturates with increasing intensity, so quadratic media possess the properties required for STS formation [6]. The absence of wave collapse in quadratic media has been demonstrated theoretically [7,8], and Malomed *et al.* [8] showed that with a positive (i.e., self-focusing) nonlinearity, strictly stable STS exist if the quadratic medium has anomalous dispersion at both the fundamental and harmonic frequencies.

Experimental evidence of solitons in quadratic nonlinear media has only begun to appear in the last few years. Stationary spatial solitons have been generated in two [9] and one [10] transverse spatial dimensions. Di Trapani *et al.* recently reported the formation of temporal solitons via the cascade nonlinearity [11]. However, to date spatiotemporal solitons in either quadratic or cubic nonlinear media have not been observed.

Here we show experimentally that STS can be produced in a quadratic medium. The effective third-order nonlinearity is produced by the cascade $\chi^{(2)}{:}\chi^{(2)}$ process. We use angular dispersion to create large and negative group-velocity dispersion (GVD) for both fundamental and harmonic pulses [11], which facilitates matching of the characteristic dispersion, diffraction, and nonlinear lengths. The self-focusing and nonlinear phase shifts balance the effects of diffraction and negative GVD to produce a soliton in one transverse spatial dimension and time. Contrary to the expectations of some previous workers [11] but consistent with others [8,12,13], we find that STS exist when the group-velocity mismatch (GVM) between fundamental and harmonic pulses is much larger than the input pulse duration in addition to when GVM ≈ 0.

The equations that govern the interaction between fundamental and harmonic electric fields E_1 and E_2 propagating in the z direction and assumed constant in the x direction are

$$\left(\frac{\partial}{\partial z} + \frac{iL_{\rm NL}}{4L_{\rm DS1}}\frac{\partial^2}{\partial t^2} + \frac{iL_{\rm NL}}{4L_{\rm DF1}}\frac{\partial^2}{\partial y^2}\right)E_1 = iE_1^*E_2e^{i\Delta kz} + i2\pi(n_2I_0)\frac{L_{\rm NL}}{\lambda}[|E_1|^2 + 2|E_2|^2]E_1\,,$$

$$\left(\frac{\partial}{\partial z} + \frac{L_{\rm NL}}{L_{\rm GVM}}\frac{\partial}{\partial t} + \frac{iL_{\rm NL}}{4L_{\rm DS2}}\frac{\partial^2}{\partial t^2} + \frac{iL_{\rm NL}}{4L_{\rm DF2}}\frac{\partial}{\partial y^2}\right)E_2 = iE_1E_1e^{-i\Delta kz} + i4\pi(n_2I_0)\frac{L_{\rm NL}}{\lambda}[2|E_1|^2 + |E_2|^2]E_2\,.$$

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E_1 and E_2 are in units of the initial value of the peak fundamental field E_0, which is related to the initial peak intensity I_0 by $I_0 = \sqrt{\frac{\varepsilon}{\mu}}\,|E_0|^2/2$. n_2 is the Kerr nonlinear index, and Δk is the wave-vector mismatch between fundamental and harmonic fields. The diffraction, dispersion, and nonlinear lengths characterizing the pulse interaction are $L_{\mathrm{DF}} = k\omega_0^2/2$, $L_{\mathrm{DS}} = 0.322\tau_0^2/|\beta^{(2)}|$, and $L_{\mathrm{NL}} = n\lambda/(\pi\chi^{(2)}E_0)$, with λ the fundamental wavelength. L_{DS2} is the dispersion length at the second-harmonic frequency, for example. Time t is measured in units of the input-pulse duration τ_0, while positions z and y are measured in units of L_{NL} and y-dimension beam waist ω_0, respectively. $\beta^{(2)}$ is the magnitude of the group-velocity dispersion, and ω_0 is the radius of the beam waist. Finally, $L_{\mathrm{GVM}} = c\tau_0/(n_{1g} - n_{2g})$ with n_{1g} (n_{2g}) the group index at the fundamental (harmonic) frequency.

To model the pulse propagation, we numerically solved the coupled wave equations in one transverse dimension. This is referred to as a $(1 + 1 + 1)$D medium, where the elements of the triplet are the transverse space, time, and longitudinal space dimensions, respectively. We used a symmetric split-step beam-propagation method to solve the coupled equations. A fourth-order Runge-Kutta algorithm solves the nonlinear propagation step in the time domain, and the dispersive-and-diffractive propagation step is solved in the frequency (and spatial frequency) domain, after a two-dimensional Fourier transform. The temporal profile and transverse intensity distribution of the initial fundamental pulse are taken to be Gaussians. The results of the numerical simulations will be compared to experimental results below.

A pulse is expected to evolve to a STS when the GVD balances the nonlinear phase modulation and diffraction balances the self-focusing. The goal of the experimental arrangement is therefore to match L_{DF}, L_{DS}, and L_{NL}. Experiments were performed with a 1-cm-long $LiIO_3$ crystal cut for type-I phase matching. $LiIO_3$ was chosen because (i) it has a large value of $\chi^{(2)}$ and (ii) its large GVM implies sufficiently large GVD to observe soliton formation in a reasonably short propagation length with achromatic phase matching [14]. Pulses of duration 110 fs and energy up to 1 mJ at a wavelength of 795 nm are produced by a Ti:sapphire regenerative amplifier. The pulses traverse a diffraction grating, a 1:1 telescope, and a cylindrical lens. The optical system creates large negative GVD by dispersing wavelengths in the x direction, while focusing the beam in the y direction (Fig. 1). The beam waist of 40 μm (full width at half maximum) in the y direction at the entrance face of the $LiIO_3$ crystal produces a diffraction length $L_{\mathrm{DF}} = 2.9$ mm, about one-third of the length of the crystal. The beam size in the x direction is $\sim$4 mm, which corresponds to a walk-off length much longer than the crystal, so spatial walk-off in the x-z plane is negligible. Anomalous GVD of magnitude $|\beta^{(2)}| = 1350$ fs^2/mm produced by the grating [14] yields

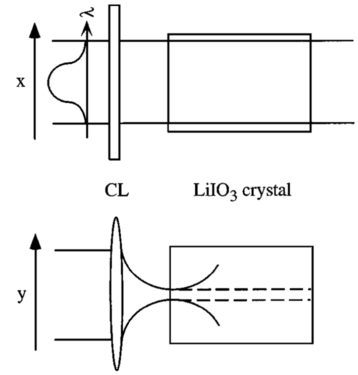

FIG. 1. Schematic of the experimental geometry. Propagation is along the z direction. A diffraction grating disperses different wavelengths in the x-z plane, as indicated in the top panel. A cylindrical lens (CL) focuses the beam in the y direction. The diffraction length in the x direction is much greater than the length of the crystal; different wavelength components propagate in slightly different directions to achieve achromatic phase matching. The dashed lines in the lower panel suggest the beam propagation when the spatiotemporal soliton is formed.

$L_{\mathrm{DS}} = 2.9$ mm for 110-fs pulses. For an ordinary type-I interaction, the GVM corresponds to a delay between pulses of 2.3 ps within one dispersion length. The angular dispersion from the grating is designed to produce achromatic phase matching, in which GVM ≈ 0. By rotating the crystal 180° about the z axis from the achromatic phase-matching orientation, the GVM can be doubled, to 4.6 ps. L_{NL} can be adjusted by varying the input pulse energy or the phase mismatch, which determine $(n_2)_{\mathrm{eff}}$. After the crystal, an identical combination of grating and lenses cancels the GVD and recollimates the beam. The fundamental pulses entering and exiting the crystal are monitored by a charge-coupled-device (CCD) camera, an autocorrelator, and a diode-array spectrometer. The beam profile of the second-harmonic pulses is also measured with the CCD camera.

A minor shortcoming of the experimental setup is the introduction of residual higher-order dispersion by the gratings and lenses used to create and nullify the large GVD. Third- and fourth-order dispersion arising from aberrations of the doublet lenses [15] currently in the experiment combine to limit the shortest pulses that can be measured accurately. Without the $LiIO_3$ crystal, 110-fs input pulses are broadened to 130 fs owing to the uncompensated higher-order dispersion. This implies that pulses shorter than $\sim$80 fs cannot be measured accurately and will produce apparent pulse durations of $\sim$80 fs.

Initially the achromatic phase matching was used to achieve GVM ≈ 0. With the $LiIO_3$ crystal rotated about the optical axis by 90° to eliminate $\chi^{(2)}$ effects, the pulse interacts with the medium through the Kerr nonlinearity alone. At higher intensities, nonlinear dispersive propagation is observed as expected. With an intensity of 50 GW/cm^2 the pulse broadens from 110 to $\sim$200 fs after propagating through the crystal. The beam profile at the output face of the crystal exhibits the effects of diffraction, broadening to $\sim 80\ \mu$m.

With the crystal oriented on the self-focusing side of phase matching ($\Delta k = k_{2\omega} - 2k_\omega < 0$), the output pulse duration and beam profile depend strongly on intensity, as expected theoretically. $\Delta kL < 0$ produces a nonlinear phase shift $\Delta\Phi^{\mathrm{NL}} > 0$. For low input intensities ($<1\ \mathrm{GW/cm^2}$) dispersive propagation is again observed along with diffraction. When the intensity of the input pulse reaches a threshold value ($10-50\ \mathrm{GW/cm^2}$) that depends weakly on ΔkL, the output pulse begins to narrow temporally and the beam waist simultaneously decreases in the y direction. Pulse intensity autocorrelations and beam profiles are shown in Fig. 2 for $\Delta kL = -80\pi$ and intensities near and well above threshold. The corresponding simulation results for the pulse temporal envelope and beam profile are shown in Figs. 2(c) and 2(d) and agree reasonably well with the experiments. Although suppression of spatial and temporal broadening is the most obvious manifestation of the STS, the observed deep modulation of the output spectrum [Fig. 3(a)] also agrees with theory [Fig. 3(b)]. Finally, the second-harmonic beam size also decreases dramatically with intensity, which confirms the mutual trapping of the fundamental and harmonic beams.

Qualitatively similar trends are observed over wide ranges of phase mismatch (approximately $-1000\pi < \Delta kL < -30\pi$) and intensity ($10 < I < 80\ \mathrm{GW/cm^2}$). Measurements made with $\Delta kL = -80\pi$ and $\Delta kL = -240\pi$ (Fig. 4) exemplify these trends. The pulse duration decreases from ~ 300 to ~ 80 fs, and the spectrum broadens by $\sim 50\%$ due to the nonlinear phase modulation. The measured and calculated pulse durations [Fig. 4(a)] agree qualitatively, but the quantitative agreement is not as good as that obtained for the beam waist [Fig. 4(b)]. For intensities $>50\ \mathrm{GW/cm^2}$, the discrepancies are at least partly attributable to the limitations of the measurement system; the nearly constant apparent pulse durations recorded at high intensities imply that shorter pulses are actually generated. For intensities $>50\ \mathrm{GW/cm^2}$, the beam waist in the y direction is reduced by a factor of 12 compared to the ordinary diffracting beam and by a factor of 3 compared to the input beam, in excellent agreement with calculations. The harmonic-pulse energies range from a few percent of the fundamental energy at small ΔkL to 0.1% at the largest values of ΔkL.

With $\Delta kL > 0$, the nonlinear phase shift produced in the cascade process $\Delta\Phi^{\mathrm{NL}} < 0$ and cancels the Kerr phase shift to some degree. At the highest input intensities the net nonlinear phase shift should be small and negative. With $\Delta kL > 0$ we do not observe STS at any input intensity up to $100\ \mathrm{GW/cm^2}$, which is the damage threshold of $\mathrm{LiIO_3}$. Together with the control experiments mentioned above, this demonstrates the crucial role played by the cascade process in the formation of STS. The critical power for one-dimensional self-focusing by the Kerr nonlinearity corresponds to an intensity of roughly $80\ \mathrm{GW/cm^2}$ in our experiment. Thus, all of our observations of STS occur below that power. For $\Delta kL = -80\pi$ we estimate that the cascade phase shift is 3–4 times that produced by the Kerr effect, which implies a threshold between 10 and $20\ \mathrm{GW/cm^2}$, as is found in both experiment and simulations. Of course, only the cascade part of the nonlinearity saturates.

With the crystal rotated about the optical axis by 180°, the formation of STS with large GVM can be studied. In linear propagation GVM would separate the fundamental and harmonic pulses by 4.6 ps (40 times the input-pulse duration) after one dispersion length. Under these conditions, we observe STS that are qualitatively

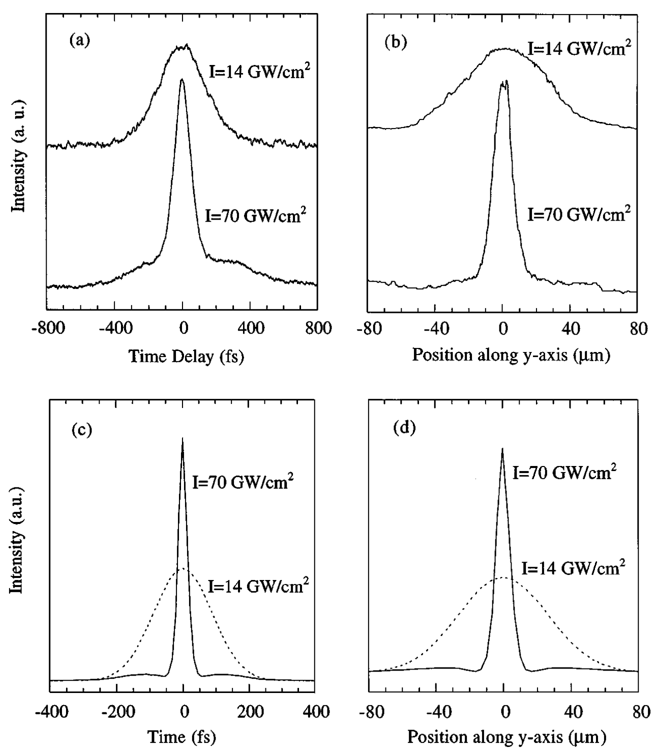

FIG. 2. Intensity autocorrelations (a) and beam profiles (b) of output pulses generated with the indicated intensities. Calculations of the pulse intensity and beam profile are shown in (c) and (d).

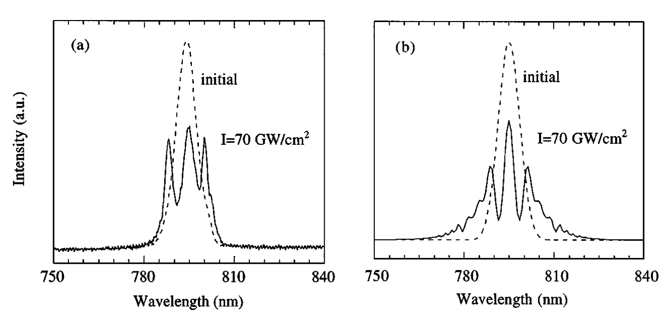

FIG. 3. Measured (a) and calculated (b) power spectrum of the STS generated at the indicated intensity.

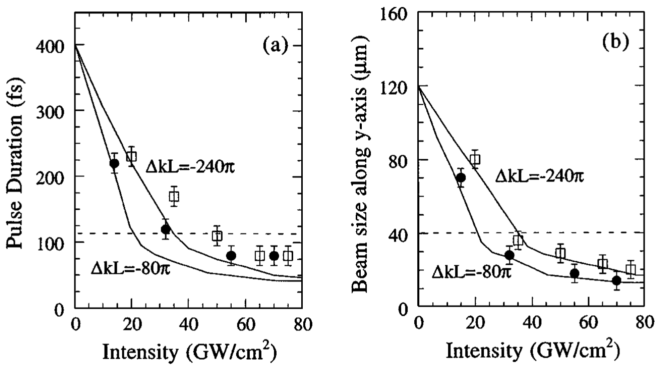

FIG. 4. Dependence of output pulse duration (a) and beam size (b) on incident intensity for the indicated values of phase mismatch. Symbols are measured points and solid lines are the results of calculations. The pulse duration and beam size incident on the crystal are indicated by the dashed lines. The filled (open) symbols correspond to $\Delta kL = -80\pi$ (-240π).

similar to those found with GVM ≈ 0. At small phase mismatch ($60\pi < |\Delta kL| < 100\pi$), the threshold intensities for formation of STS are 20%–40% higher than those observed with GVM ≈ 0. On the other hand, at large phase mismatch ($|\Delta kL| > 200\pi$), the thresholds are similar to the zero-GVM values. Typical results obtained with $\Delta kL = -240\pi$ are shown in Fig. 5. Di Trapani *et al.* [11] claimed that GVM can prevent the formation of STS by splitting the interacting pulses before the mutual trapping mechanism takes effect, and so only investigated temporal solitons formed with GVM ≈ 0. Our measurements contradict this claim. Torner *et al.* have shown that solitons theoretically can form in the presence of walk-off (GVM) in the case of one transverse spatial (temporal) dimension [13].

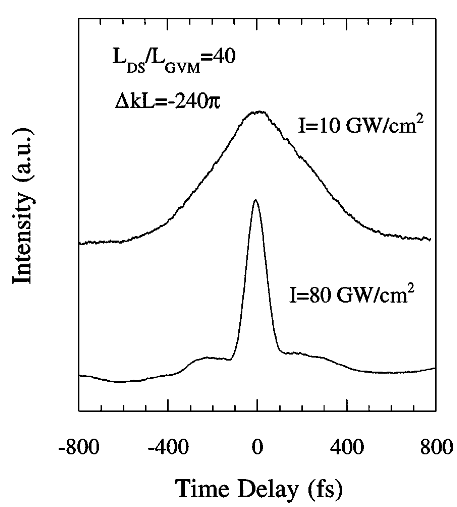

FIG. 5. Intensity autocorrelations measured just below and well above threshold for STS formation with GVM equal to 40 times the input-pulse duration.

In conclusion, we have generated stable $(1 + 1 + 1)$D spatiotemporal solitons in a quadratic nonlinear medium. The fundamental and harmonic pulses trap each other spatially and temporally, overcoming the effects of diffraction (and walk-off) and group-velocity dispersion. Here we reach conclusions about soliton formation by using a medium 3 times longer than the characteristic interaction length; direct confirmation of stable propagation over longer distances, along with studies of the evolution of input pulses to solitons, are planned. We expect that spatiotemporal solitons will find numerous applications in science and technology in the future.

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[1] For example, *Solitons*, edited by M. Lakshmanan (Springer-Verlag, Berlin, 1988).
[2] This area is reviewed in J. Opt. Soc. Am. B **14**, 2950–3253 (1997). Theoretical and experimental achievements in the important photorefractive nonlinear systems are reviewed by M. Segev and G. Stegman in Phys. Today **51**, No. 1, 42 (1998).
[3] Y. Silberberg, Opt. Lett. **15**, 1282 (1990).
[4] E. A. Kuznetsov, A. M. Rubenchik, and V. E. Zakharov, Phys. Rep. **142**, 105 (1986); J. J. Rasmussen and K. Rypdal, Phys. Scr. **33**, 481 (1986).
[5] H. J. Bakker, P. C. M. Planken, L. Kuipers, and A. Lagendijk, Phys. Rev. A **42**, 4085 (1990).
[6] Y. N. Karamzin and A. P. Sukhorukov, JETP Lett. **41**, 414 (1976).
[7] A. A. Kanashov and A. M. Rubenchik, Physica (Amsterdam) **4D**, 122 (1981).
[8] B. A. Malomed, P. Drummond, H. He, A. Berntson, D. Anderson, and M. Lisak, Phys. Rev. E **56**, 4725 (1997).
[9] W. E. Torruellas, Z. Wang, D. J. Hagan, E. W. VanStryland, G. Stegeman, L. Torner, and C. R. Menyuk, Phys. Rev. Lett. **74**, 5036 (1995).
[10] R. Schiek, Y. Baek, and G. I. Stegeman, Phys. Rev. E **53**, 1138 (1996).
[11] P. Di Trapani, D. Caironi, G. Valiulis, A. Dubietis, R. Danielus, and A. Piskarskas, Phys. Rev. Lett. **81**, 570 (1998).
[12] D. Mihalache, D. Mazilu, B. A. Malomed, and L. Torner, Opt. Commun. **152**, 365 (1998).
[13] L. Torner, C. R. Menyuk, and G. I. Stegeman, J. Opt. Soc. Am. B **12**, 889 (1995); L. Torner, D. Mazilu, and D. Mihalache, Phys. Rev. Lett. **77**, 2455 (1996).
[14] O. E. Martinez, IEEE J. Quantum Electron. **25**, 2464 (1989).
[15] W. E. White, F. G. Patterson, R. L. Combs, D. F. Price, and R. L. Shepherd, Opt. Lett. **18**, 1343 (1993).