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Self-Trapping of Spatially and Temporally Incoherent White Light in a Photochemical Medium

Jihua Zhang, Kailash Kasala, Abhitej Rewari, and Kalaichelvi Saravanamuttu*

Department of Chemistry, McMaster University, 1280 Main Street West, Hamilton ON L8S 4M1, Canada

Received October 13, 2005; E-mail: kalai@mcmaster.ca

The natural diffraction—spatial broadening—of light is suppressed in certain nonlinear photosensitive materials in which a beam induces its own narrow waveguide and propagates as a self-trapped optical mode. Self-trapped beams and their extraordinary particle-like interactions have been studied for over four decades in different nonlinear optical (NLO) materials.[1,2] Self-written fibers in polymers have been attributed to similar self-trapping processes.[3] Studied predominantly with (temporally and/or spatially coherent) laser light, self-trapped beams were assumed to be a strictly coherent species until 1997 when Mitchell and Segev discovered that white light—which is spatially and temporally incoherent—self-trapped in a NLO photorefractive crystal.[4] Their finding stimulated entirely new lines of theoretical research,[2,5,6] but experimental advances have been limited by the properties of materials normally used for self-trapping studies; the fast photoresponses of conventional NLO materials render them sensitive to the femtosecond-scale random phase fluctuations characteristic of white light. This causes the beam to break-up, disrupting self-trapping. The only strategy that overcomes this limitation resorts to extremely low (nW) optical powers; this forces a slow response from the medium, which "sees" only the time-averaged (smoothed) intensity profile of white light and allows self-trapping of the rapidly fluctuating broadband beam.[4] In this paper, we report that self-trapping of white light is enabled by a photo*chemical* reaction. We demonstrate that a beam of incoherent white light self-traps by initiating polymerization in an organosiloxane gel.[7,8] Self-trapping is possible because the photoresponse time of this medium, which is determined by the *inherently* slow rate of a polymerization reaction, exceeds the femtosecond time scale by several orders of magnitude.[3]

We used a gel consisting of methacrylate-substituted siloxane oligomers sensitized to visible wavelengths with a free-radical photoinitiator (Supporting Information).[7,8] White light from an incandescent quartz−tungsten−halogen lamp was focused onto the entrance face of the gel contained in a transparent cuvette. The intensity profile of the beam in the gel was imaged onto a CCD camera. Figure 1a shows that the input focused beam at the entrance face has a width (fwhm) of 45.0 μm and relative peak intensity of 38.4%. The beam broadens to 268.5 μm with an intensity of 15.4% after propagating 6.0 mm in the gel (Figure 1b). We monitored the temporal evolution of this diffracted beam; images of the beam profile are presented in Figure 1, and corresponding temporal plots of peak intensity and beam widths are presented in Figure 2b. Within seconds, the initially broad and diffuse beam undergoes severe distortions (Figure 1c−f) before self-focusing into a single peak (Figure 1g), which remains narrow and intense over a prolonged period of time (Figure 1g−j). The contrast between the self-trapped and initial diffracted profiles of the beam is striking; the former shows a 5.4-fold decrease in width and a >6.5-fold increase in relative intensity relative to the latter. The self-trapped beam widens slightly over time with a corresponding decrease in intensity (Figure 1k−m) but did not return to its original diffracted state for as long as it was monitored (>2000 s).

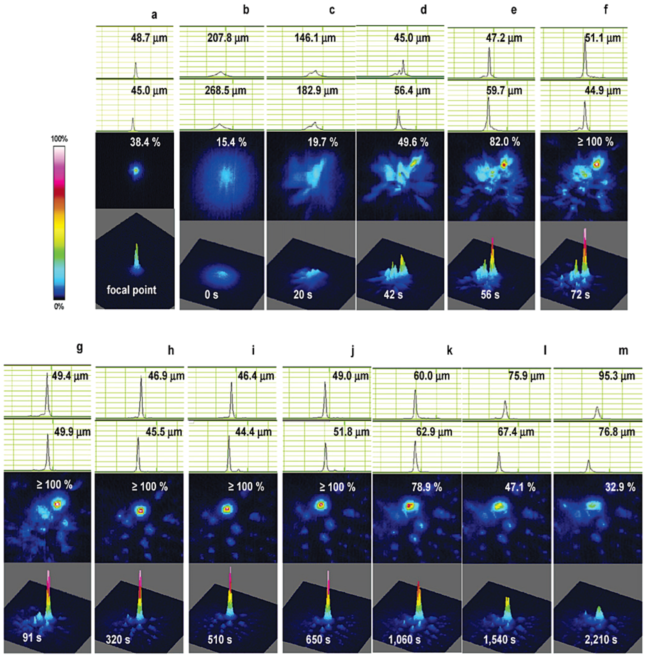

Figure 1. From top to bottom: 1-D horizontal, 1-D vertical (scale = 600 μm/division), 2-D and 3-D intensity profiles of the white light beam at (a) the entrance face and (b−r) during self-trapping at a propagation distance of 6.0 mm in the organosiloxane gel. The fwhm widths for the horizontal and vertical profiles (μm), the relative peak intensity (%), and the time (s) are indicated. The color graph represents the scale for relative peak intensity (see Table 1 and video in Supporting Information).

The self-trapping process begins when the input beam initiates free-radical polymerization of methacrylate substituents at the entrance face. The polymerization rate, which is proportional to optical intensity,[9] varies according to the (time-averaged) intensity profile of the input beam (Figure 1a); it maximizes at the (most intense) axial region and decays radially from this point. Because refractive index changes due to polymerization are spatially localized in the gel (with at least 150 nm spatial resolution[8]), the intensity profile of the 45.0 μm wide beam induces a corresponding gradient in refractive index—this is a lens. Instead of diffracting as it propagates from the entrance face, the beam encounters a lens and self-focuses into the gel; at its new focal point, it induces a second lens to focus even further. Its reciprocal lensing and focusing actions proceed along the propagation axis, causing significant narrowing of the beam width from 268.5 to 44.9 μm with a corresponding increase in intensity from 15.4 to ≥100%, which corresponds to saturation of the CCD camera (Figure 1f).

The suppression of diffraction during self-focusing is clearly seen in Figure 1d−h as intensity from peripheral regions of the beam is channeled into the growing higher index (higher intensity) peak.

10.1021/ja056838+ CCC: $33.50

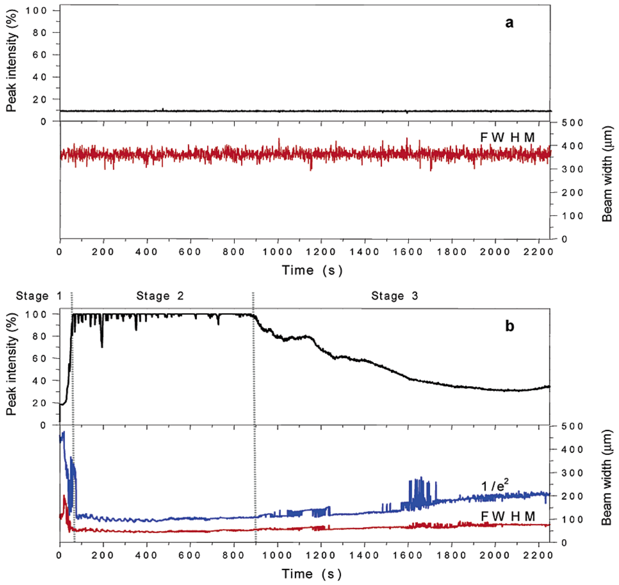

Figure 2. (a) Temporal plot of the relative peak intensity (black) and fwhm width (red) of a diffracted beam in an organosiloxane medium without photoinitiator. (b) Temporal plots of relative peak intensity (black), fwhm (red), and $1/e^2$ (blue) widths. Three distinct stages of the process—self-focusing, self-trapping, and waveguide broadening—are marked as Stage 1, Stage 2, and stage 3, respectively.

The brief appearance of multiple peaks at early times in Figure 1d−f is probably due to local inhomogeneities in photoinitiator concentration or temperature fluctuations due to the exothermic polymerization process. However, the highest intensity peak rapidly dominates, and the entire beam of white light confines itself within this single maximum in 72 s (Figure 1f). Self-focusing is observed as a large and rapid increase in peak intensity and decrease in beam width in Stage 1 of Figure 2b.

Refractive index changes induced by the beam along its propagation axis create a channel waveguide with a gradient in refractive index that maximizes at the axis. The beam, which is confined to the higher-index axial region, remains focused and intense during propagation (Figure 1f−k). We measured the self-trapped width of the beam at different propagation distances; the widths were 47.9, 49.9, and 49.0 μm at distances of 5.0, 6.0, and 7.0 mm, respectively. These values correspond closely to the input beam width (45.0 μm) and show that the beam maintains a focused and narrow profile (i.e., it is self-trapped) throughout propagation. During Stage 2 (Figure 2b) of self-trapping, the beam is stable, sustaining an intensity maximum of $\geq$100% and beam widths between 44.9 and 52.7 μm for a prolonged period of time (728 s).

Over time, all methacrylate groups in the path of the beam are consumed by polymerization. (Previous studies showed that polymerization leads to a maximum refractive index change of 0.006.[7]) This means that refractive index changes saturate and the waveguide develops a uniform refractive index profile. Instead of being confined to the higher-index core, optical intensity is now distributed over the entire cross-section of the waveguide, causing a gradual increase in beam width and corresponding decrease in intensity. As the beam widens, polymerization is initiated at its peripheral regions, which broadens the waveguide and, in turn, the width of the self-trapped beam (Figure 1k−m). In Figure 2b, Stage 3 of self-trapping is seen as a gradual decrease in intensity and a corresponding increase in beam width. The white light beam remains self-trapped and does not return to its original diffracting state (Figure 1b) for as long as we monitored the experiment. At 2210 s, the beam had an intensity of 32.9% and a width of 76.8 μm (Figure 1m). This represents a 2.14-fold increase in intensity and 3.5-fold decrease in width relative to its original diffracted state (Figure 1a).

To prove that the entire spectrum of white light (and not simply a narrow band of wavelengths) self-traps, spectra of the self-trapped beam were acquired at a propagation distance of 6.0 mm. The spectral range of the original diffracted beam, which ranges from 500 to 800 nm, remained unchanged throughout self-trapping (Supporting Information). Self-trapping of white light was also observed in the organosiloxane gels over a range of optical powers (0.20−1.64 mW limited only by the power range of the QTH lamp). Over 100 self-trapping experiments have been carried out in the organosiloxane gels with a success rate of >90%. (Experiments failed when the organosiloxane gel developed macroscopic inhomogeneities that could scatter visible light.) To confirm that self-trapping of white light was directly due to free-radical polymerization, a control experiment was carried out in a nonphotosensitized organosiloxane medium. Temporal plots of relative peak intensity and beam width in Figure 2a show that, in the absence of free-radical polymerization, the diffracting beam remains broad and weak and does not self-trap.

Because excited states in a hot incandescent source decay independently of one another, the emitted white light is composed of a distribution of optical modes—a pattern of intensity speckles[4]—which randomly fluctuates on the femtosecond time scale. We have shown that such a spatially and temporally incoherent light beam self-traps as a single entity due to the *noninstantaneous* changes in refractive index caused by a polymerization reaction; that is, the self-induced waveguide supports multiple optical modes at visible wavelengths (500−800 nm). Refractive index changes of the waveguide should also be sufficient to guide the longer telecommunication NIR wavelengths (1.3 and 1.55 μm). Our photochemical approach makes it possible to systematically vary properties such as rate and magnitude of refractive index change and photosensitivity through the chemical composition of the system (e.g., methacrylate concentration). This introduces the possibility of studying self-trapping of incoherent light under material parameters that are not available to conventional nonlinear optical materials, whose photoresponse is determined by higher-order dielectric susceptibility tensors.

Acknowledgment. We thank C. Giardino for preliminary experiments, Prof. J. Preston for discussions, and Ciba Specialty Chemicals, Canada for donation of the photoinitiator. This project was funded by NSERC, CFI, OIT, and McMaster University.

Supporting Information Available: Video of temporal evolution of beam profile during self-trapping. Table of beam parameters corresponding to Figure 1. Spectra of self-trapped beam. This material is available free of charge via the Internet at http://pubs.acs.org.

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JA056838+