Optical power limiting and stabilization using a two-photon absorbing neat liquid crystal in isotropic phase

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This letter reports on a neat liquid crystal that incorporates a two-photon-absorbing chromophore in its structure. The nonlinear optical properties including two-photon-induced fluorescence spectrum and decay behavior, as well as the effective two-photon absorption coefficient of this liquid crystal in its isotropic phase are presented. Fairly good optical limiting and stabilization performance with this nonlinear material have been achieved by using ~815-nm and ~5-ns laser pulses. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1588364]

Optical power limiting[1] and optical stabilization[2] are of considerable interest for researchers. These two techniques are highly useful (i) for protection of optical devices from intense light-induced damages, and (ii) for stabilizing the power or intensity of an intense light beam to reduce temporal fluctuations and noises. One of the best approaches to reach these goals is based on two- or three-photon absorption in a proper nonlinear absorbing material.[2] Compared to various other mechanisms and approaches (such as reverse saturable absorption, induced refraction, and induced thermal aberration), a multiphoton-absorption-based approach exhibits the advantages of negligible linear attenuation losses and fast temporal response, and therefore can be employed for optical stabilization purposes and remain a good optical quality of the transmitted signals.

So far, a considerable number of studies on multiphoton-absorption-based optical limiting and stabilization have been reported by using dye solutions,[3–5] dye-doped solid matrices,[6,7] and dye-doped liquid crystals.[8] In all these cases, the effectiveness of optical limiting or stabilization performance depended on (i) the two (or three)-photon-absorption cross-section values of the dye molecules and (ii) the dye concentration values for a given optical thickness of nonlinear absorbing samples. In practice, the dye concentration values are usually limited to 10^{-2}–10^{-1} M in solution phase[3,4] as well as in doped solid matrices[6,7] due to dye segregation at higher concentrations. Therefore, a specific type of organic nonlinear material is desirable that exhibits a much higher effective concentration of absorbing centers than the aforementioned values.

In this letter we report a neat liquid crystal, consisting of two-photon-absorbing chromophores, which therefore possesses a much higher effective concentration of nonlinear absorbing centers. The chromophore core was obtained by carrying out the Heck reaction for tri(4-bromophenyl)amine and 4-acetoxystyrene.[9] After hydrolysis and followed by esterification with 3,4,5-tridodecyloxybenzoic acid catalyzed by dicyclohexylcarbodiimide and 4-N,N-dimethylaminopyridine in dry tetrahydrofuran, the desired compound, tri[4′-(3,4,5-tridodecyloxybenzoyloxy)-stilbenyl]amine was obtained. The chemical structure of this compound is shown in Fig. 1. This compound is a discotic liquid crystal at room temperature, and changes to the isotropic liquid phase at 87 °C. Upon cooling from the isotropic phase, the columnar phase starts to appear at 80 °C and this mesophase will maintain at room temperature for at least 72 h. The details of synthesis and mesophase behavior will be reported elsewhere.

The linear absorption spectrum of a 10-μm-thick liquid crystal film at room temperature is also shown in Fig. 1. The sample film was prepared by heating a liquid crystal drop between two glass slides to ~100 °C and then pressing the two slides to form a film, in which the thickness could be controlled via a proper spacer between the slides. The spectral curve shown in Fig. 1 was obtained 1 h after the heating ended and the optical loss from two cover slides was subtracted. It is seen in Fig. 1 that there is no linear absorption in the very broad spectral range from 500 to 1100 nm. However, upon excitation with an intense IR pulsed laser beam of wavelength about 800 nm, a frequency-upconverted and visible fluorescence could be readily observed from the liquid

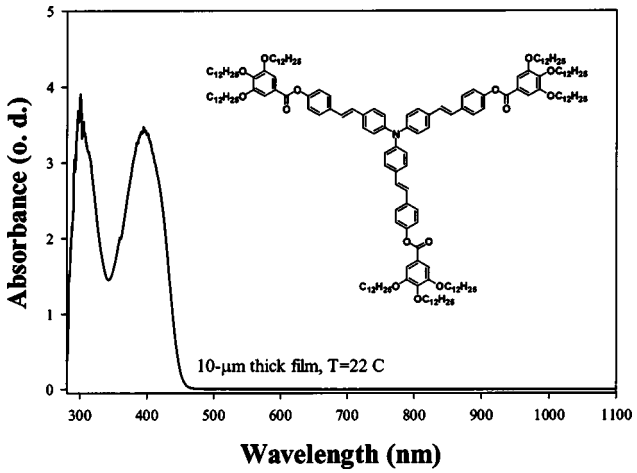

FIG. 1. Linear absorption spectrum of a 10-μm-thick liquid crystal film at room temperature. The chemical structure of this sample is shown in the right-top corner.

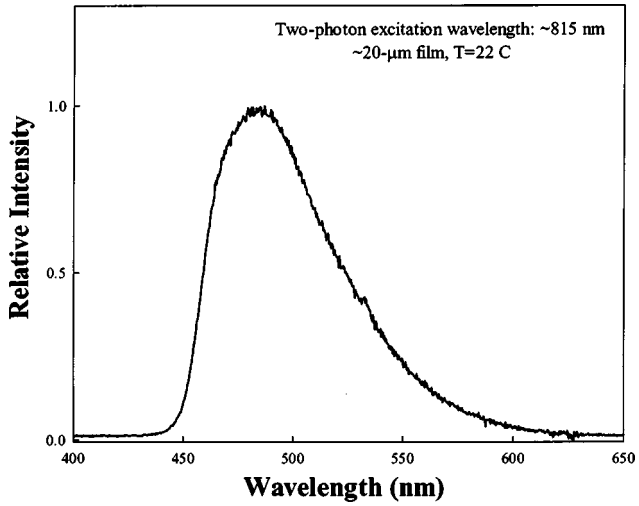

FIG. 2. Two-photon-induced fluorescence spectrum of a 20-μm-thick liquid crystal film at roomtemperature, excited by ~815-nm and ~5-ns laser pulses.

crystal sample either in its film form at room temperature or in the isotropic liquid phase at temperature $\geqslant 87\,^{\circ}\mathrm{C}$. This frequency-upconverted emission is due to two-photon absorption (TPA) of the input IR laser radiation. As an example, Fig. 2 shows the TPA-induced fluorescence spectrum of an ~20-μm-thick liquid crystal film at room temperature, obtained 1 h after cooling down from the isotropic liquid phase. The excitation beam was provided by a pulsed dye laser system with the following output characteristics: wavelength ~815 nm, pulse duration ~5 ns, beam divergence angle ~1 mrad, and repetition rate 10 Hz.

The dynamic behavior of TPA-induced fluorescence was studied by using a high-speed streak camera (C5680-22 from Hamamatsu) in conjunction with an ultrashort IR excitation pulse source that was a Ti:sapphire laser/amplifier system producing ~140-fs laser pulses of wavelength ~790 nm at a repetition rate of 1 kHz. The measured TPA-induced fluorescence decay following an excitation laser pulse is shown in Fig. 3 by a solid-line curve, which could be well fitted with a double exponential curve (dashed line) with two decay

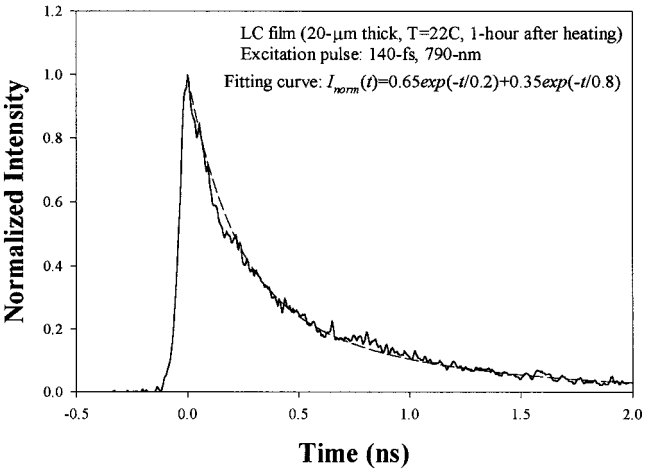

FIG. 3. Measured two-photon-induced fluorescence decay curve (solid line) excited by ~790-nm and ~140-fs laser pulses. The dashed line is a double exponential decay curve with two decay constants of 0.2 and 0.8 ns respectively.

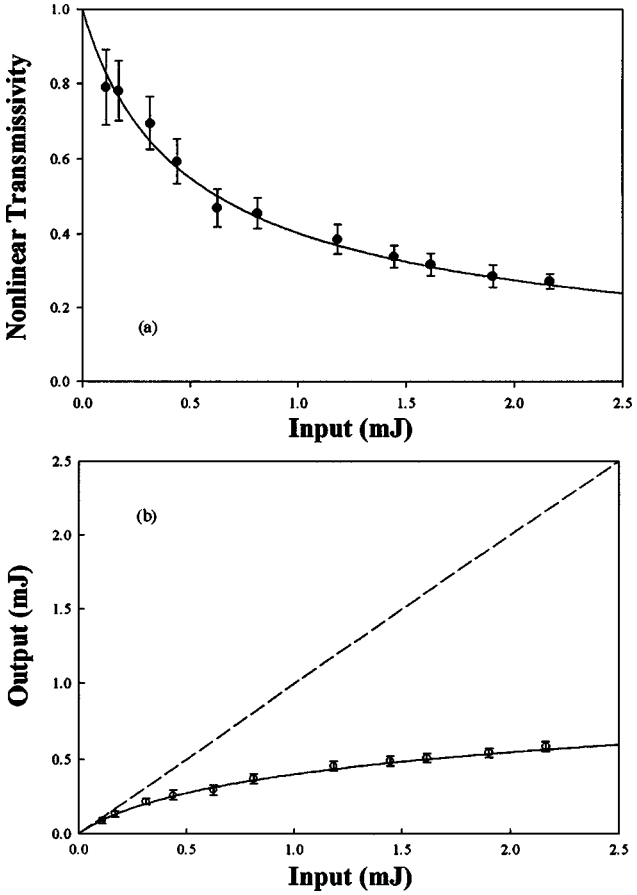

FIG. 4. (a) Measured nonlinear transmissivity data as a function of the input energy; the thick solid line is the best fitting curve with $\beta = 6.25$ cm/GW. (b) Measured output energy versus input; the thin solid line is the best fitting curve with the same β value.

constants of 0.2 and 0.8 ns. The resolution of the streak camera in this scale is ~25 ps.

From the viewpoint of application for optical power limiting and stabilization, the nonlinear medium should be highly transparent for a weak input optical beam with an appropriate wavelength, but becomes more absorptive at higher intensity input. At room temperature, if the thickness of our liquid crystal film is greater than 0.5 mm, the linear transmission drops down significantly even for a weak input beam due to strong scattering loss inside the thick liquid crystal layer in the columnar hexagonal (D_h) phase. The scattering losses for a 0.1-mm-thick film (at room temperature) at different wavelength potions were measured to be 4.9 dB (at 500 nm), 3.8 dB (at 550 nm), 2.8 dB (at 600 nm), 1.8 dB (at 700 nm), and 0.9 dB (at 800 nm), respectively. For this reason, our optical limiting performance was conducted by using a 1-cm-long quartz cell filled with the liquid crystal sample that was kept at $\sim 100\,^{\circ}\mathrm{C}$. Therefore, the sample was linearly transparent in the spectral range of 500 to 1100 nm; that is, without scattering loss. The pulsed IR beam of ~815-nm wavelength and ~5-ns pulse duration from the dye laser source was passed through an ~1.5-mm diaphragm and then focused by an $f = 20$-cm lens into the center of the 1-cm cell. The measured nonlinear transmissivity and the output pulse energy as a function of the input pulse energy are shown in Figs. 4(a) and 4(b), respectively. The input laser pulse energy was controlled through a polariza-

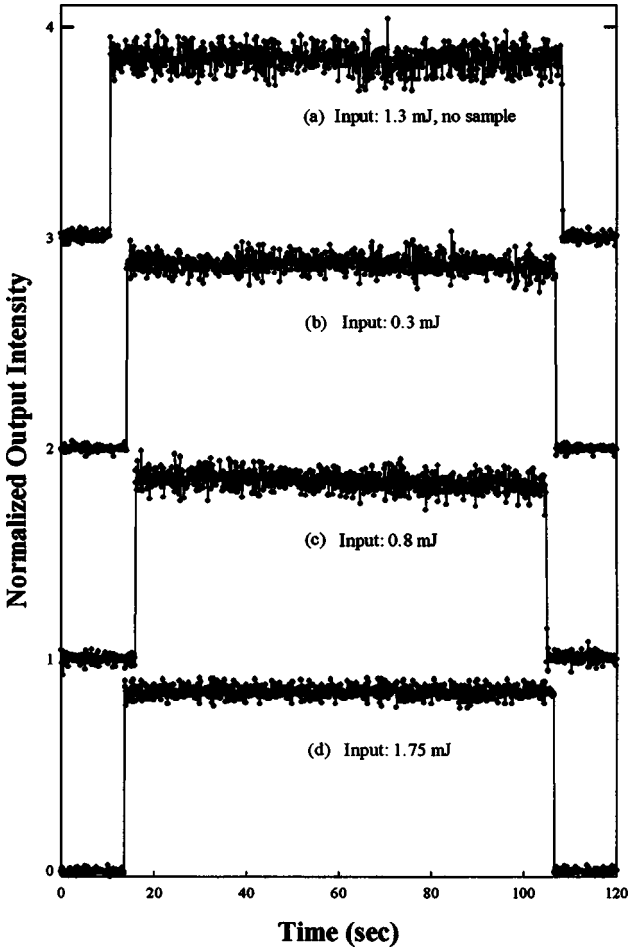

FIG. 5. Measured temporal energy (intensity) fluctuation for input pulses at the average level of (a) 1.3 mJ, and for output pulses at the average input levels of (b) 0.3 mJ, (c) 0.8 mJ, and (d) 1.75 mJ.

tion prism with a variable attenuation ratio. According to the basic theory of TPA,[1] the nonlinear transmissivity can be expressed as

$$T(I_0)=[\ln(1+\beta l I_0)]/(\beta l I_0), \tag{1}$$

where I_0 is the input light intensity, l is the thickness of the sample, and β is the TPA coefficient of the nonlinearly absorbing medium. In Fig. 4(a), the thick solid-line curve is a theoretical curve obtained by the use of Eq. (1) with the best fitting value of $\beta=6.25$ cm/GW, while in Fig. 4(b), the thin solid-line curve is a theoretically predicted curve using the same β value. From Fig. 4(b), one can see that when the input energy levels varied from 0.2 to 2.0 mJ (10 times increase), the transmitted energy changed only from 0.15 to 0.55 mJ (3.7 times increase). This is a typical optical limiting behavior.

On the other hand, from Fig. 4(b) it is also seen that if the input energy (or intensity) level fluctuates within a range, for instance, from 1.5 to 2.0 mJ, the output level fluctuates within a much small range; that is, from 0.49 to 0.55 mJ. In this case the relative fluctuation values for the input and output are $\Delta_0=0.29$ and $\Delta'=0.12$, respectively. Based on this observation, one can use a high-TPA medium to effectively reduce the intensity fluctuation and to stabilize the optical power at a constant level. As an experimental example, the optical stabilization behavior of our 1-cm-long liquid crystal sample (at $\sim 100\,^{\circ}$C) is demonstrated in Fig. 5. In Fig. 5, the temporal fluctuation of the input energy (at an average level of 1.3 mJ) is shown by curve (a), whereas the output fluctuation characteristics at different average input levels are shown by curves (b), (c), and (d), respectively. All data were obtained by using a gated integrator (Model 4422 from EG&G), working at gate width of 10 ns and in conjunction with a fast photodiode detector. The measured time range for each curve was about 90–100 s, and each measured point corresponded to one of the 10-Hz laser pulses. It is noted that the temporal fluctuation of the input laser pulses is not dependent on their average energy levels because of the use of a polarization prism as the variable attenuator. The relative fluctuation for the input signals was measured to be $\Delta_0 \approx 0.33$ [see curve (a) in Fig. 5], whereas the relative fluctuation for the output signal (at 1.75-mJ input level) was measured to be $\Delta' \approx 0.15$ [see curve (d) in Fig. 5]. This is a typical optical stabilization behavior.

In conclusion, we have demonstrated fairly good optical limiting and stabilization performance using a neat liquid crystal as an intrinsic two-photon-absorbing medium. This type of materials can also be employed for other studies such as multiphoton pumped and frequency-upconverted lasing, IR-visible image conversion, and three-dimensional data storage. As another example, the two- and three-photon pumped frequency-upconversion lasing has been recently demonstrated in a neat liquid dye system that also exhibits very high effective dye concentration.[10]

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