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# Femtosecond filament amplification in liquids

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**ABSTRACT** We study the femtosecond filamentation in a liquid amplifying medium, sulphorhodamine 640 in a methanol solvent. In the presence of population inversion, the energy of the filament increases while its diameter expands because of peak intensity clamping.

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## 1 Introduction

Much attention has been given recently to the process of femtosecond filamentation. Laser pulses undergoing filamentation exhibit remarkable features, which make them very interesting for applications. The pulse duration shortens significantly, reaching nearly the single cycle limit. In the best cases [1–3], the pulse spectrum broadens until it spans the entire visible [4], and the beam quality improves to single transverse mode [5, 6]. A particularly attractive feature is the fact that these effects correspond to a self-action, which does not require exacting alignment procedures. One of the key issues is the amount of energy that can be carried by a filament. Most studies so far show that for an incident power well beyond the critical power for filamentation, the beam decomposes into a multifilamentary pattern, each of the subfilaments carrying the same amount of energy. Although this multiple pattern can be organized by suitable amplitude and phase modulation of the incident pulse [7–9], it nevertheless represents a major limiting effect in many applications. We have examined the possibility of increasing the filament energy by generating filaments in an amplifying medium. In this scheme, the amplifying medium consists of a transparent nonlinear host responsible for filamentation by optical Kerr self-focusing and plasma defocusing, while a dopant is responsible for broadband fluence amplification. Since the intensity is clamped by multiphoton ionization of the medium [10, 11], single filament amplification should correspond to an increase of the size of the emerging filament. A recent experiment in a solid has demonstrated the possibility to increase the filament energy while avoiding multiple filament formation [12]. However, the amplified filament reached the threshold for irreversible damage of the host crystal, preventing further detailed studies of filament beam expansion and useful applications. We have therefore investigated filament amplification in a dye in solution, where damage is less an issue.

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## 2 Experimental set-up

The scheme of the experiment is shown in Fig. 1. The laser pulse is produced in an amplified CPM (colliding-pulse mode-locked) laser chain [13, 14]. The incident pulse has a duration of 80 fs and a maximum energy per pulse of 3 mJ. The filament is formed in a Bethune [15] 7 cm long cell, which contains $1.6 \times 10^{-5}$ M/l of sulphorhodamine 640 dye diluted in methanol. The dye cell is put at a Brewster angle to minimize energy losses and to avoid laser effects inside the cell due to internal reflections on the cell faces. The Bethune cell containing the liquid is pumped homogeneously with a 10 ns pulse at 532 nm (obtained by second harmonic generation of a Nd:YAG laser).

It is well known that the broadband gain profile of a dye in solution makes it possible to obtain very short pulses. Filamentation in transparent liquids and in un-pumped dye solutions has been also demonstrated previously [16]. Figure 2 shows a 1 cm portion of the side image of the luminescence due to a filament in a passive (un-pumped) cell containing the sulphorhodamine 640 solution in methanol. The single filament has an average radius of 30 μm, it starts 4 cm after the front entrance of the cell and is maintained over a distance of 3 cm until the end of the cell. The energy of the un-amplified

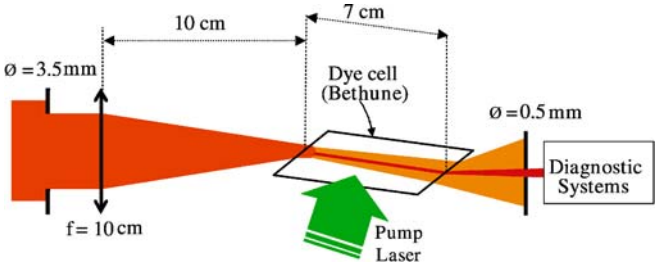


**FIGURE 1** Experimental set-up: The incident laser pulse at 610 nm (incident from the left) is focused into a dye cell, which can be pumped with a ns pulse at 532 nm. A filament is formed in the cell. Its characteristics with and without amplification are examined at the exit window plane

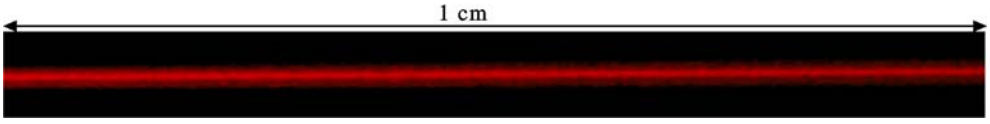


**FIGURE 2** Portion of the side view of the filament in an un-pumped cell with sulphorhodamine 640 in methanol solvent. The image corresponds to the fluorescence of the dye excited by the filament

filament is 10 μJ. The filament energy was measured with a calibrated photodiode (spectral range 200–1100 nm) at the exit of the dye cell. The filament core was filtered with a $\varphi = 0.5$ mm spatial filter placed 5 cm after the exit face of the cell, as shown in Fig. 1.

## 3 Intensity clamping

Filament formation and amplification can be easily distinguished from the usual pulse amplification in the dye cell through its important spectrum broadening, as shown in Fig. 3. Figure 3a (dash-dotted line) shows the spectrum of the amplified pulse emerging from the dye cell in the absence of filamentation. This is obtained by reducing the incident pulse power below the value necessary for filamentation or by removing the focusing lens. Figure 3a (continuous line) shows the spectrum of the amplified pulse emerging from the dye cell in presence of filamentation. Figure 3b shows the spectrum of the filament in the passive and active cell. From Fig. 3b it is evident that there is no significant variation between the spectrum of an un-pumped and amplified filament. This indicates that the $B$-integral ($B(r,t) = \int n_2 I(r,t)\,\mathrm{d}z$) responsible for self-phase modulation is the same. We conclude, therefore, that the amplification of the filament does not change the filament peak intensity. This is in agreement with all previous studies of femtosecond laser filamentation, which revealed a saturation of the peak intensity in a filament (intensity clamping).

## 4 Measurement of energy gain inside the filament

We now investigate the energy gain in the filamentation. The energy gain is defined as the ratio between the energy of the amplified and un-pumped filament. The set-up of the energy measurement is described in the Sect. 2. The results of the measurement are shown in Fig. 4. We can see that the energy inside the filament core grows with the pump energy until gain saturation occurs around 10–20 mJ of green pump energy. We measure a saturated fluence amplification of the filament by a factor 9, with a maximum energy of 90 μJ in a single filament.

## 5 Measurement of filament diameter expansion

The visualization of the beam cross section directly on the exit face of the cell is difficult because of the large Brewster angle between the cell and the beam and because of the light scattered at the exit surface. The filament diameter was measured indirectly, by recording the profile of the beam cross section at different distances beyond the cell exit face, along the propagation direction. The 1-D filament profiles, shown in Fig. 5e and f, correspond to beam cross sections recorded in the Brewster plane, where the beam diameter is

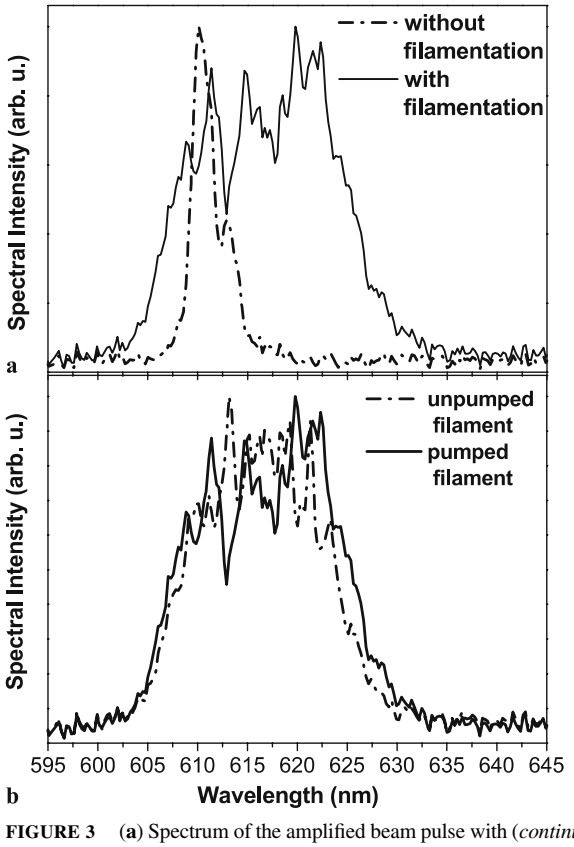


**FIGURE 3** (**a**) Spectrum of the amplified beam pulse with (*continuous line*) and without (*dash-dot line*) filamentation. (**b**) Spectrum of the pulse for an amplified (*continuous line*) and an un-amplified (*dash-dot line*) filament

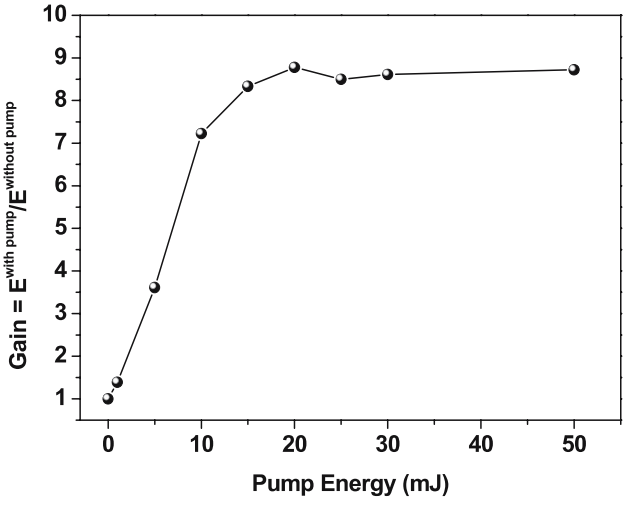


**FIGURE 4** Behavior of the energy gain inside the filament as function of the green pump energy

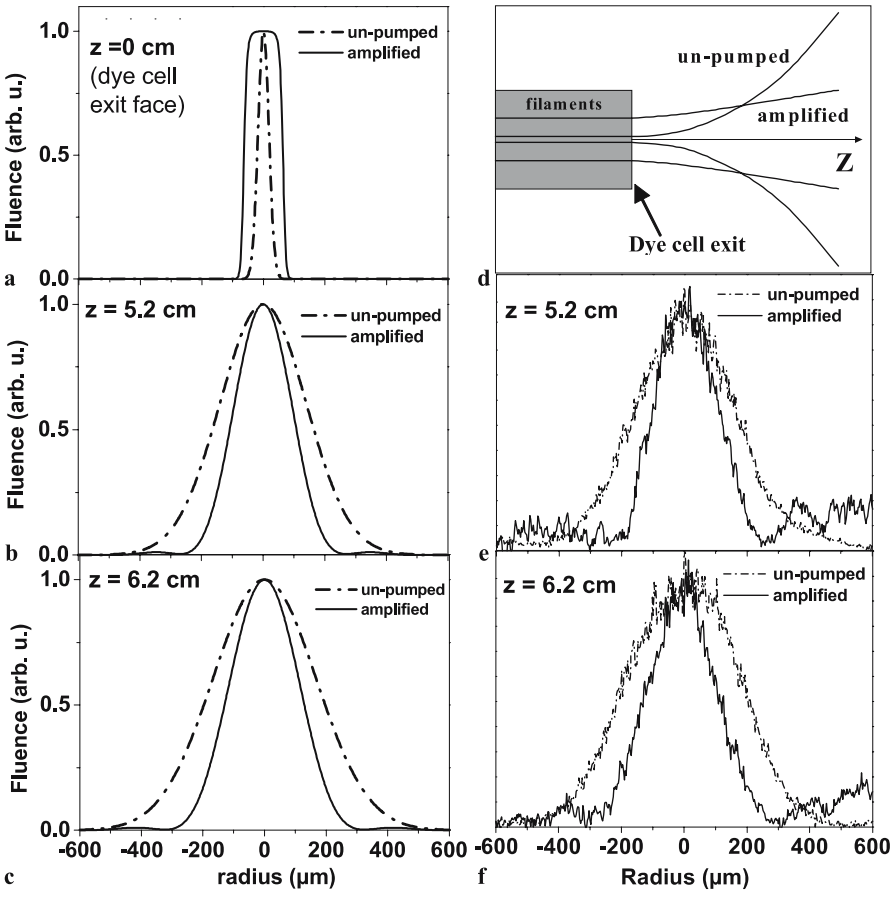


**FIGURE 5** (**a**) shows the assumed Gaussian profile of the un-pumped filament (*dash-dotted line*) and the theoretical profile of the filament (*continuous line*) in presence of population inversion in the cell as computed by the model of saturated fluence gain specified in (3). Figures (**b**), (**c**), (**e**), (**f**) show a comparison between the computed filament profiles (**b** and **c**) and the respectively profiles measured with a CCD camera (**e** and **f**), after propagation on 5.2 cm (**b** and **e**) and 6.2 cm (**c** and **f**). The profiles of the un-pumped filament are shown in *dash-dotted line* while the profiles of the amplified filament are shown in *continuous line*. Figure (**d**) shows qualitatively the behavior of the diffracted beams beyond the cell exit face

not distorted by refraction at the exit surface, both for the amplified and non-amplified filaments. The measurements were performed at two distances $z = 5.2$ cm and $z = 6.2$ cm from the cell, for different energies of the green pump laser. The measured profiles of the amplified filaments are independent of the pump energy beyond 10 mJ. In order to extract the filament size at $z = 0$ (the exit plane of the cell), we first consider the filament without amplification. For the un-amplified filament we assume a Gaussian fluence profile:

$$F(r) = F_m \exp\left(-\frac{2r^2}{w_0^2}\right), \qquad (1)$$

where $F_m$ is the peak fluence in the filament. We apply to the filament propagation the linear spatial diffraction law: $w(z) = w_0\sqrt{1+z^2/b^2}$, where $w(z)$ is the beam diameter at a point $z$ (half-width at $1/e^2$), $w_0$ the waist of the beam and $b = \pi w_0^2/\lambda$ is the Rayleigh length. From the measurement of the beam diameter at two different distances beyond the dye cell, $w_1 = w(z_1)$ and $w_2 = w(z_2)$, one can calculate the Rayleigh length $b$ and then the waist $w_0$ of the beam by resolving the 2nd degree equation:

$$(K^2+4)(g^2-1)b^2 - 2Kd(g^2+1)b + d^2(g^2-1) = 0, \qquad (2)$$

where $K = \pi\Delta^2\lambda d$, $\Delta^2 = w_2^2 - w_1^2$ and $g = w_2/w_1$ are coefficients determined by the measurement. With this method, we find that the filament radius on the exit surface of the dye cell is 30 μm. (See the dash-dotted line of Fig. 5a and Table 1.)

Because of intensity clamping and saturation of the gain, it is reasonable to introduce for the amplified filament a phenomenological profile of the form:

$$F'(r) \propto F_m \exp\left(-\frac{2r^2}{w_0^2}\right) \times \left[1+\kappa\exp\left(-\frac{2r^2}{w_0^2}\right)\right]^{-1}, \qquad (3)$$

where $\kappa$ is a saturation parameter adjusted to reproduce the results of measurements. This supergaussian-like profile is used as an initial condition in a linear 2-D code of propagation beyond the exit surface. The propagated beam profiles after 5.2 and 6.2 cm of propagation are compared with the profiles measured with the CCD camera at the same distances. The numerical results reproduce well the measured values of the filament diameter for a value of $\kappa = 1000$. With such value

| Distance in cm | $Z = 0$ (exit cell face) | $Z = 5.2$ | $Z = 6.2$ |
|---|---|---|---|
| Radius in μm (un-amplified) | 30 (calculated) | 330 (measured) | 390 (measured) |
| Radius in μm (un-amplified) | 130 (calculated) | 220 (measured) 213 (calculated) | 245 (measured) 256 (calculated) |

**TABLE 1** Comparison between measured and calculated beam width at two distances $Z = 5.2$ cm and $Z = 6.2$ cm for an un-amplified filament and a filament amplified to saturation (pump energy = 20 mJ). The beam width at $Z = 0$ is extracted from diffraction theory (see text)

of $\kappa$, we find that the filament size on the exit surface is increased from $w = 30\,\mu$m (un-pumped value) to a saturated value of 130 μm (see Fig. 5b, e, c, f and Table 1). By means of the phenomenological model described by (3), we can say that a filament can bear more energy if amplified, but because of the intensity clamping, which saturates the intensity peak in the core of the filament, its size expands resulting in a larger diameter (factor 4) than if not amplified. This is illustrated by a continuous line in Fig. 5a.

## 6 Pulse duration inside the filament

One of the interesting aspects of filamentation is the self-compression down to near the single cycle limit [1–3]. Combined with filament amplification, it opens the prospect to obtain the necessary short pulses in the visible that are required to produce X-UV attosecond pulses. We have, therefore, examined the potential of dye amplification of filament in the context of very short pulses.

A direct measurement of the pulse duration after amplification in the dye cell proved difficult. Although evidence for pulse splitting and pulse shortening was obtained from autocorrelation measurements, no reliable results could be obtained. We can, nevertheless, obtain some information on the pulse duration from an analysis of the filament spectrum and the gain.

We can estimate the pulse shortening in the case of the filament amplification. Let $I$, $w$ and $\Delta t$ be the intensity, radius and duration of the initial (un-amplified) filament and $I'$, $w'$, $\Delta t'$ the intensity, radius and duration of the filament in presence of population inversion inside the dye cell then, if $G$ represents the energy gain inside the filament, we should have

$$\frac{I'}{I} = \left(\frac{\Delta t}{\Delta t'}\right)\left(\frac{w}{w'}\right)^2 G = \left(\frac{\Delta t}{\Delta t'}\right) R \approx 1\,, \tag{4}$$

because the peak intensity does not change with the energy of the pump laser, as justified in paragraph 3.

The measured value of the quantity $R \equiv (w/w')^2 G$ becomes a constant $R \approx 0.5$ at high green pump energies (see Fig. 6). Therefore, from (4) we conclude that the pulse duration inside the amplified filament is further shortened by no more than a factor of two. This is a further temporal compression, with respect to the self-compression by filamentation,

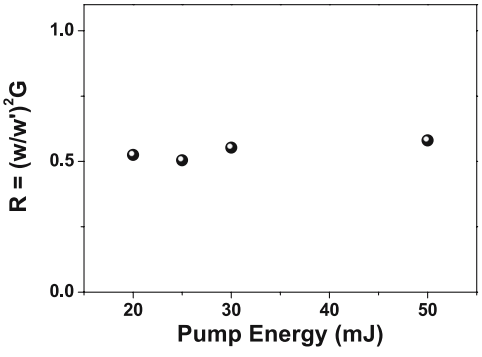


**FIGURE 6** Behaviour of the measured quantity $R$ (see the text for the definition of $R$) for 20 mJ, 25 mJ, 30 mJ and 50 mJ of energy of the green pump. The quantity $R$ has a saturated value around 0.5 in presence of high pump energy

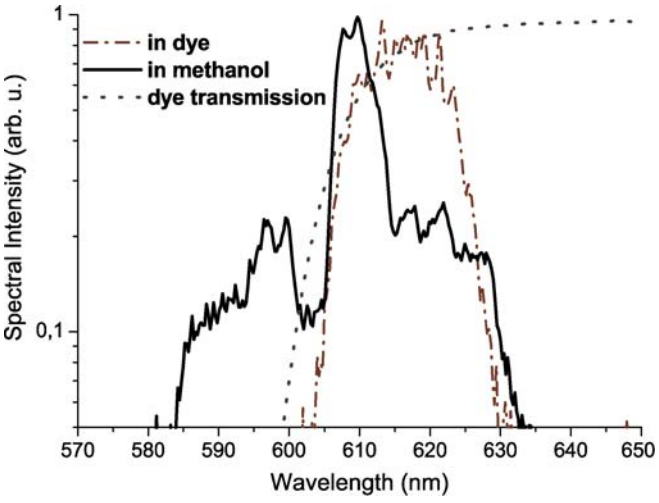


**FIGURE 7** Spectrum of the filament in sulphorhodamine 640 in methanol solvent (*dash-dotted line*) and in pure methanol (*continuous line*). In *dotted line* is shown the transmission coefficient of 25 mm of dye solution ($1.6 \times 10^{-5}$ Mol/l)

and is due to the combination of filament energy amount by amplification and size filament expansion processes.

We also note that the spectrum of the filament is limited by re-absorption by the dye. This is shown in Fig. 7 where we compare the spectrum of the un-amplified filament in methanol with and without sulphorhodamine 640 dye. It clearly shows that the blue side of spectrum broadening due to filamentation is absorbed by the dye. From the bandwidth of the amplified filament spectrum (about 20 nm, see Fig. 3b) one can estimate a minimum filament pulse duration in the dye solution of $\sim$ 15 fs. This value is the best we could obtain by the pulse temporal self-compression due to the filamentation combined to the further compression due to the filament amplification and expansion processes.

## 7 Conclusions

In this work we have demonstrated that it is possible to amplify the energy of a single femtosecond filament from 10 μJ up to 90 μJ by propagating it in an amplifying dye solution. We have also demonstrated that the filament diameter self-adjusts by expanding its diameter by a factor of four. These results indicate that the energy and the diameter of a filament are not fixed quantities as it was claimed previously [5].

We have also obtained evidence that the amplification process does not compromise the self-compression of the pulse inside a filament. In fact the analysis done in paragraph 6 shows that under amplification the filament pulse even can further be shortened by a factor of two because of energy gain, filament size expansion and peak intensity clamping. However, because of re-absorption by the dye, the emerging pulse duration does not self-compress below 15 fs in the present system. It would be interesting to test other systems such as amplifying liquids with a large Stokes shift between absorption and gain, or high pressures gases with a broad gain profile.

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## REFERENCES


1 C.P. Hauri, W. Kornelis, F.W. Helbing, A. Heinrich, A. Couairon, A. Mysyrowicz, J. Biegert, U. Keller, Appl. Phys. B **79**, 673 (2004)
2 A. Couairon, M. Franco, A. Mysyrowicz, J. Biegert, U. Keller, Opt. Lett. **30**, 2657 (2005)
3 A. Couairon, J. Biegert, C.P. Hauri, W. Kornelis, F.W. Helbing, U. Keller, A. Mysyrowicz, J. Mod. Opt. **53**, 75 (2006)
4 J. Kasparian, M. Rodriguez, G. Méjean, J. Yu, H. Wille, R. Bourayou, S. Frey, Y.-B. André, A. Mysyrowicz, R. Sauerbrey, J.-P. Wolf, L. Wöste, Science **301**, 61 (2003)
5 K.D. Moll, A.L. Gaeta, G. Fibich, Phys. Rev. Lett. **90**, 203 902 (2003)
6 B. Prade, M. Franco, A. Mysyrowicz, A. Couairon, H. Buersing, B. Eberle, M. Krenz, D. Seiffer, O. Vasseur, "Spatial Mode Cleaning by Femtosecond Filamentation", to be presented at CLEO/QELS 2006 conference
7 G. Méchain, A. Couairon, M. Franco, B. Prade, A. Mysyrowicz, Phys. Rev. Lett. **93**, 035 003 (2004)
8 V.P. Kandidov, N. Aközbek, M. Scalora, O.G. Kosareva, A.V. Nyakk, Q. Luo, S.A. Hosseini, S.L. Chin, Appl. Phys. B **80**, 267 (2005)
9 H. Schroeder, J. Liu, S. Chin, Opt. Express **12**, 4768 (2004)
10 J. Kasparian, R. Sauerbrey, S.L. Chin, Appl. Phys. B **71**, 877 (2000)
11 A. Becker, N. Aközbek, K. Vijayalakshmi, E. Oral, C.M. Bowden, S.L. Chin, Appl. Phys. B **73**, 287 (2001)
12 J. Philip, C. D'Amico, G. Chériaux, A. Couairon, B. Prade, A. Mysyrowicz, Phys. Rev. Lett. **95**, 163 901 (2005)
13 A. Migus, A. Antonetti, J. Etchepare, D. Hulin, A. Orszag, J. Opt. Soc. Am. B **4**, 584 (1985)
14 M.M. Murnane, R.W. Falcone, J. Opt. Soc. Am. B **8**, 1573 (1988)
15 D.S. Bethune, Appl. Opt. **11**, 1897 (1981)
16 H. Schroeder, S.L. Chin, Opt. Commun. **234**, 399 (2004)