Above-millijoule super-continuum generation using polarisation dependent filamentation in atoms and molecules

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Abstract: We demonstrate for the first time that input polarisation control inducing one single filamentation is a very robust technique to accurately control the filamentation dynamics enhancing throughput energy of the supercontinuum generation up to 1.2 millijoule. Reaching the above-millijoule regime opens the way to post-compression of multi-terawatt laser pulses.

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References and links

- 1. T. Pfeifer, C. Spielmann, and G. Gerber, "Femtosecond X-ray science," Rep. Prog. Phy. 69, 443-505 (2006).
- E. Goulielmakis, Yakovlev VS, Cavalieri AL, Uiberacker M, Pervak V, Apolonski A, Kienberger R, Kleineberg U, Krausz F, "Attosecond Control and Measurement: Lightwave Electronics," Science 317, 5839, 769-775 (2007).
- A. Zaïr, Holler M, Guandalini A, Schapper F, Biegert J, Gallmann L, Keller U, Wyatt A. S., Monmayrant A, Walmsley I. A., Cormier E, Auguste T, Caumes JP, Salieres P, "Quantum Path Interferences in High-Order Harmonic Generation," Phys. Rev. Lett. 100, 14, 143902 (2008).
- Eckle P, Smolarski M, Schlup P, Biegert J, Staudte A, Schoffler M, Muller HG, Dorner R, Keller U, "Attosecond angular streaking," Nature Physics 4, 7, 565-570 (2008).
- Morlens AS, Gautier J, Rey G, Zeitoun P, Caumes JP, Kos-Rosset M, Merdji H, Kazamias S, Casson K, Fajardo M, "Submicrometer digital in-line holographic microscopy at 32 nm with high-order harmonics," Opt. Lett., 31, 21, 3095-3097 (2006).
- Goulielmakis E, Koehler S, Reiter B, Schultze M, Verhoef AJ, Serebryannikov EE, Zheltikov AM, Krausz R, "Ultrabroadband, coherent light source based on self-channeling of few-cycle pulses in helium," Opt. Lett. 33, 13, 1407-1409 (2008).
- Hentschel M, Kienberger R, Spielmann C, Reider GA, Milosevic N, Brabec T, Corkum P, Heinzmann U, Drescher M, Krausz F, "Attosecond metrology," Nature 414, 509-513 (2001).
- Sansone G, Benedetti E, Calegari F, Vozzi C, Avaldi L, Flammini R, Poletto L, Villoresi P, Altucci C, Velotta R, Stagira S, De Silvestri S, Nisoli M, "Isolated single-cycle attosecond pulses," Science 314, 443 (2006).
- Paul PM, Toma ES, Breger P, Mullot G, Auge F, Balcou P, Muller HG, Agostini P, "Observation of Train of Attosecond Pulses from High Harmonic Generation," Science 292, 1689-1692 (2001).
- 10. P. Agostini and L. F. DiMauro, "The physics of attosecond light pulses," Rep. Prog. Phys. 67, 813 (2004).
- Telle HR, Steinmeyer G, Dunlop AE, Stenger J, Sutter DH, Keller U, "Carrier-envelope offset phase control: A novel concept for absolute optical frequency measurement and ultrashort pulse generation," App. Phys. B 69, 327 (1999).
- Nisoli M, DeSilvestri S, Svelto O, Szipocs R, Ferencz K, Spielmann C, Sartania S, Krausz F, "Compression of high-energy laser pulses below 5 fs," Opt. Lett. 22, 522-524 (1997).
- Hauri CP, Kornelis W, Helbing FW, Heinrich A, Couairon A, Mysyrowicz A, Biegert J, Keller U, "Generation of intense carrier-envelope phase-locked few-cycle laser pulses through filamentation," App. Phys. B 79, 673-677 (2004).

- Kosareva O. G., Panov N. A., Uryupina D. S., Kurilova M. V., Mazhorova A.V., Savelev A.B., Volkov R.V., Kandidov V.P., Chin S.L., "Optimization of a femtosecond pulse self-compression region along a filament in air," Appl. Phys. B 91, 35 (2008).
- Couairon A., Biegert J., Hauri C. P., Kornelis W., Helbing F. W., Keller U., Mysyrowicz A., "Self-compression of ultrashort laser pulses down to one optical cycle by filamentation," J. Mod. Opt., 53, 75-85(2006)
- Guandalini A, Eckle P, Anscombe M, Schlup P, Biegert J, Keller U, "5.1 fs pulses generated by filamentation and carrier-envelope phase stability analysis," J. Phys. B: Atomic Molecular and Optical Physics 39 S257-S264 (2006).
- Zaïr A, Guandalini A, Schapper, Holler M, Biegert J, Gallmann L, Keller U, Couairon A, Franco M, Mysyrowicz A, "Spatio-temporal characterization of few-cycle pulses obtained by filamentation," Optics Express 15, 9, 5394-5404 (2007).
- 18. Skupin S, Stibenz G, Berge L, Lederer F, Sokollik T, Schnurer M, Zhavoronkov N, Steinmeyer G, Phys. Rev. E 74, 056604 (2006).
- G. Stibenz, N. Zhavoronkov, G. Steinmeyer, "Self-compression by femtosecond pulse filamentation: Experiments versus numerical simulations," Opt. Lett. 31, 2, 274-276 (2006).
- Prade B, Franco M, Mysyrowicz A, Couairon A, Buersing H, Eberle B, Krenz M, Seiffer D, Vasseur O, "Spatial mode cleaning by femtosecond filamentation in air," Opt. Lett. 31, 2601-2603 (2006).
- Lange HR, Chiron A, Ripoche JF, Mysyrowicz A, Breger P, Agostini P, "High-Order Harmonic generation and quasiphase matching in Xenon using self-guided femtosecond pulses," Phys. Rev. Lett. 81, 8, 1611-1613 (1998).
- Liu W, Petit S, Becker A, Akozbek N, Bowden CM, Chin SL, "Intensity clamping of a femtosecond laser pulse in condensed matter," Opt. Commun. 202, 1-3, 189-197 (2002).
- Becker A, Akozbek N, Vijayalakshmi K, Oral E, Bowden CM, Chin SL, "Intensity clamping and re-focusing of intense femtosecond laser pulses in nitrogen molecular gas," App. Phys. B. 73, 3, 287-290 (2001).
- A. Couairon and A. Mysyrowicz, "Femtosecond filamentation in transparent media," Physics Reports 441, 2-4, 47-189 (2007).
- C. Iaconis and I. A. Walmsley, "Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses," Opt. Lett. 23, 792794 (1998).
- Main P, Strickland D, Bado P, Pessot M, Mourou G, "Generation of Ultrahigh Peak Power Pulses by. Chirped Pulse Amplification," IEEE J. Quantum Electron. 24, 2, 398-403 (1988).
- S. Petit, A. Talebpour, A. Proulx, S.L. Chin, "Polarization dependence of the propagation of intense laser pulses in air," Opt. Commun. 175, 4-6, 323-327 (2000).
- N. Aközbek, M. Scalora, C.M. Bowden and S.L. Chin, "White light continuum generation and filamentation during the propagation of ultra-short laser pulses in air," Opt. Commun. 191, 353 (2001).
- H. Schroeder, S.A. Hosseini, Q. Luo, S.L. Chin, "Self-steepening is an abrupt process," Opt. Commun. 266, 1, 302-306 (2006).
- M. V. Ammosov, N. B. Delone, and V. P. Krainov, "Tunnel ionization of complex atoms and atomic ions in an electromagnetic field," Sovietic Physics: Journal of Experimental and Theoretical Physics 64, 1191 (1986).
- M. Kolesik, J.V. Moloney and E.M. Wright, "Polarization dynamics of femtosecond pulses propagating in air," Phys. Rev. E 64 046607(2001)
- 32. G. Fibich and B. Ilan, "Multiple Filamentation of Circularly Polarized Beams," Phys. Rev. Lett. 89 (1), 1-4, 013901(2002).
- 33. G. Fibich and B. Ilan,"Self-focusing of circularly polarized beams," Phys. Rev. E 67 (3), 1-15,036622 (2003).
- A. Trisorio, A and CP. Hauri, "Control and characterization of multiple circularly polarized femtosecond filaments in argon," Opt. Lett. 32, 1650-1652 (2007).
- 35. A. K. Dharmadhikari, J. A. Dharmadhikari, F. A. Rajgara, and D. Mathur, "Polarization and energy stability of filamentation-generated few-cycle pulses," Optics Express **16**,10, 7083-7090 (2008).
- S. N. Vlasov, V. A. Gaponov, I. V. Eremina and L. V. Piskunova, "Self-focusing of wave beams with elliptical polarization," Radiophys. Quantum Electron. 21, 4, 358-362 (1978).
- Fedorov V. Yu., Kandidov V. P., Kosareva O. G., Akozbek N, Scalora M., ChinS. L., "Filamentation of a femtosecond laser pulse with the initial beam ellipticity," Laser Physics 16, 8, 1227-1234 (2006).
- X. M. Tong, Z. X. Zhao and C. D. Lin, "Theory of molecular tunneling ionization," Phys.l Rev. A 66, 033402 (2002).
- 39. the filament visible length corresponds to an effective length defined as a series of focusing-defocusing cycles. A proper filament length estimation require terahertz measurement.
- 40. G. Fibich and N. Gavish, "Critical power of collapsing vortices," Phys. Rev. A 77, 045803 (2008).
- 41. G.P. Agrawal, "Nonlinear fiber optics", (3rd ed.). San Diego: Academic Press, 2001.
- Couairon A., Mchain G., Tzortzakis S., Franco M., Lamouroux B., Prade B., Mysyrowicz A. "Propagation of twin laser pulses in air and concatenation of plasma strings produced by femtosecond infrared filaments," Opt. Commun. 225, 177 (2003).

1. Introduction

The development of techniques for few-cycles laser pulses generation has been one of the recent achievements that opened the way to dramatic improvements in many fields of research pushing the frontier of high field physics [1, 2, 3, 4] and extreme nonlinear optics [5, 6] towards the nonadiabatic limit [7, 8, 9, 10]. These techniques together with carried envelope phase stabilisation (CEP) [11] are one of the key point for generation of extreme ultraviolet (XUV) attosecond pulses train [9] and isolated attosecond pulses [7, 8]. To produce few- cycle laser pulses the first technique was proposed by M. Nisoli and coworkers [12] using self-phase modulation in gas-filled hollow fibers enabling to produce 3.8 fs laser pulses with hundred microjoules energy throughput. To avoid fiber-laser coupling issues and maximum energy input limitation an alternative technique based on filamentation in gas was proposed and demonstrated [13, 14, 15]. It provided sub-5fs pulses with hundreds of microjoules, selecting the output spatial inner core [17], maintaining the CEP [16] and a good output beam spatial quality. All these systems make use of additional chirp compensation based on chirp mirrors that could be apparently supressed making use of filamentation self-compression [18, 19]. Practically, the filamentation is obtained by focusing a laser pulse in a gas cell, leading to its nonlinear propagation over several Rayleigh length parameter. This propagation is now understood as an equilibrium between the Kerr effect, leading to beam focusing and plasma generation inducing beam defocusing inducing a broadening of the incident pulse spectrum to more than an optical octave and a self-guiding that improves the spatial mode quality [20]. In addition, the process inproves the intensity stability due to the pulse intensity clamping that occurs inside the filament [21, 22, 23]. Over all these properties, the underlying physics of filamentation is supported by well established numerical modeling and continue to spark off interest and ideas [24]. Nevertheless, even if most of the present filamentation set-up provide sub-5 fs pulses the energy throughput still remains limited to few hundreds of microjoule. However, regarding the robustness of the filamentation technique in the gigawatt regime and the advance in laser technology to provide more and more powerfull laser pulses up to multi-terawatt regime, we think that this method could be a good candidate to export the production of few-cycles pulses to the multi-millijoule throughput energy regime.

In this work, our motivation is to generate a supercontinuum source with more than a millijoule (supra-mJ) energy throughput, increasing the input power and avoiding for multifilamentation formation. We looked for a laser macroscopic parameter easy to control and which does not depend significantly on laser fluctuations. Therefore we oriented our choice on the input laser polarisation easly controlled by an input quarter-wave plate whilst the output ellipticity can be adjusted by a properly oriented broadband quarter-wave plate to afford the supercontinuum generated. Here we present a complete experimental study of filamentation behavior with respect to the ellipticity degree of the input laser pulse. We demonstrate experimentally for the first time to our knowledge that a polarisation control is an efficient tool to increase the input power maintaining one single filament providing a supercontinuum of 300 nm bandwidth. We managed with one stage filament to carry energy throughput up to 1.2 mJ and to break the mJ limit. Besides, we highlight generic and singular behavior of the single filament generated in atomic (argon) or molecular (nitrogen) gas targets. This is the first step towards multi-millijoule supercontinuum and few-cycles pulses generation necessary to explore high flux secondary sources in high intense non-adiabatic conditions.

1.1. Supercontinuum control on input polarisation

We selected 4 mJ from our Titane: Sapphire multi-terawatt laser system that provides 805 nm, 30 fs pulses operating at 10 Hz repetition rate (Amplitude Technologies). The input (a) and output (b) laser beams were measured with SPIDER [25] and are presented in Fig. 1. In a first

step the laser input energy was attenuated to $600 \,\mu$ J and focused with a spherical silver mirror with a radius of curvature (ROC) of -3000 mm into a 2 m long cell filled with nitrogen at 1520 mbar. The output of the filamentation exhibits a broadened spectrum, with its center of gravity shifted toward shorter wavelengths due to ionisation. Collimating the output beam and selecting the inner core we obtain an energy of 200 μ J after standard chirp mirrors compensation with 200 nm large spectra. We measured reproducible 12.3 fs pulses profile (Fig. 1(b)) confirming a compression factor close to 2.5.



Fig. 1. Input (FWHM = 29.7 fs) and output filament pulse (FWHM = 12.3 fs) profiles: the filament is generated in one stage cell filled with nitrogen at atmospheric pressure.

Taking this measurement as a starting point, we fixed the cell pressure and allowed two degrees of freedom: the input energy that could be scaled from 0 to 4 mJ by a set of half-wave plate and polariser and the ellipticity degree tuned by a quarter-wave plate. The CPA[26] laser compressor is adjusted to compensate for the total matter dispersion In a first experiment we fixed the input energy to 2 mJ corresponding to input peak laser intensity of $1.1 \times 10^{14} W/cm^2$ below the targets barrier supression $(2.4 \times 10^{14} W/cm^2)$ for argon and $2.2 \times 10^{14} W/cm^2$) and enough to consider that ionisation is driven by tunnel process. The gas pressure is fixed to 1180 mbar for the argon case and 1630 mbar for the nitrogen one to generate one single filament in linear input polarisation. We compared the effect of the polarisation control on a single filament mode generated in the atomic versus molecular target. We choose these specific couple of targets because they have the same ionisation potential $(I_p(Ar) = 15.79 \, eV)$ and $I_p(N_2) =$ $15.58\,eV$), ensuring filamentation under the same ionisation regime. To probe the filamentation dynamic by polarisation control [27] we study the evolution of the output spectra, in the inner core selection, with respect to the input ellipticity degree ε . In this experiment the dynamic is ellipticity dependent, therefore we did observed that a specific inner core selection had to be done. The difference of selection was not dramatic 3-4 mm. Indeed the largest supercontinuum, the smallest inner core selection. The smallest selection define an inferior limit necesary for being independent of the ellipticity so that for this measurement, we choose the 3 mm selection and did all the measurement through it. The results are shown in Fig. 2(a) for argon and (b) for nitrogen. We observe a common behaviour for the atomic and the molecular targets: in linear polarisation ($\varepsilon = 0$) a supercontinuum up to 300 nm is generated consistent with a pulse duration below 7 fs. These spectra exhibit a long blue tail appearing as constant flat top shoulder (nitrogen case) and modulated one (argon case) from 600 nm to 400 nm. In both cases the blue shoulder corresponds to frequencies generated by Kerr effect in the self-steepening region of the pulse[28, 29]. Independently of the gas nature, changing the ellipticity degree from 0 (linear polarisation) to 1 (circular polarisation) induces a decrease of the spectra bandwidth

from 300 nm to $\approx 60 nm$. Indeed increasing the ellipticity degree we basically decrease the ionisation rate [30] and increase the critical power for collapse [31] $P_{cr}^{\varepsilon=0} < P_{cr}^{\varepsilon=1}$. As a result the self-phase modulation and the plasma formation are less efficient in circular polarisation so that to be in the same conditions of filamentation obtained in the linear polarisation case, one needs to increase the input energy.



Fig. 2. Single filament spectra dependence on the ellipticity degree for argon (a: left) and nitrogen (b: right) targets

1.2. Spatial control: Converging to single-filamentation

Assuming now that for producing a single filament it is possible to start with a higher input energy with circular polarisation than with a linear polarisation, we propose to define the maximum input energy for our experimental study in argon and nitrogen. To do so we explore the possibility to reduce multi-filamentation formation[32, 33] by controlling the ellipticity degree and the input energy. For this second experiment we maintained the pressure to the previous values and we fixed the input energy to 3.6 mJ in order to induce multi-filamentation in argon and in nitrogen for a linear polarisation. Thus we increase the ellipticity degree to study the multi-filamentation spatial profile behavior. The results are shown in Fig. 3(a) for argon target and (b) for nitrogen one. Note that the nitrogen molecules are randomly oriented and that at $1.1 \times 10^{14} W/cm^2$, only single ionisation occurs. Under the MO-ADK theory, X. M.Tong and coworkers [38] demonstrated for linear polarisation, that nitrogen single ionisation rate is three times lower than argon one. As a result, for the same input energy we produce 2-3 filaments in nitrogen and more than 5 in argon.

The results show a similar behavior in both targets: increasing the ellipticity degree form linear polarisation ($\varepsilon = 0$) to circular polarisation ($\varepsilon = 1$) the number of multi-filaments is reduced until converging to a single filament mode. In the inner core selection of the single filament produced by circular light, we observe supercontinuum generation of $\approx 300 nm$. In order to correctly measure the energy in this supercontinuum we define the selected output inner core as performed in [17]. Note that in this experiment where we change the ellipticity and adjust the input energy in order to always produce the same single filament, the dynamic is maintained therefore it is independent on the ellipticity. We define the inner core again resulting as a selection of 3 mm for all the measurements. This selection done, we measured 0.8 mJ for argon target while an energy up to 1.2 mJ is obtained for nitrogen. We also notice that with a initial circular polarisation the filament output pointing and energy stability is better as recorded in [34, 35]. This shows that controlling the laser input pulse polarisation one can force the multi-



Fig. 3. Multi-filament spatial profile dependence on the ellipticity degree for argon (a: left) and nitrogen (b: right) targets. The focal point is shown at the top and present an elliptic spatial shape which explain multi-filaments distribution.

filamentation to converge to a single filament containing supra-mJ supercontinuum generation up to 300 nm for a fixed input energy.

1.3. Molecular and atomic behavior

Generating filament in atomic and molecular targets, we observed general behavior on the spectral and spatial frame by the polarisation control. Looking in details differences appear between argon and nitrogen filament generation. Even if for both targets the nonlinearty could be comparable, the ionisation contribution remains different. In linear polarisation we notice that the arrangement of the multi-filaments distribution presents a different transversal alignment from argon to nitrogen. To understand we first measure the focal spot in air with low energy to avoid nonlinear effects. The spatial focus distribution shown in Fig. 3 presents a clear elliptic spatial shape that is responsible for spatial anisotropic propagation and explains the alignment of the multi-filaments [36, 37]. However it does not explain the clear difference of the alignment orientation from argon to nitrogen. To understand this spatial orientation we first supposed that an aligment of the molecules in the leading edge of the pulse might provide an additional anisotropy. To probe this hypothesis we generated double-filamentation in nitrogen in linear polarisation and with a half-wave plate we modify the orientation of the linear polarisation. We did not see any effect on the double-filamentation orientation. Indeed for short pulses (tens femtoseconds) one can consider that the time scale whilst the alignment is longer than hundred of femtoseconds, therefore it cannot be responsible of the observed discreapancy. The only process left to explain the differences observed between the argon and the nitrogen filament alignment must be then the ionisation process. The key point to find the origin of the different behaviour is on the particular electronic structure of the nitrogen molecule. The ionisation

does not occur along the molecular axis but along the orientation of the valence electron orbital, meaning that the ionisation depends on the molecular symmetry. For the particular case of nitrogen, the valence electron occupies a σ_g state which is an elongated orbital. This particular electronic structure induces an anisotropic electronic density after the ionisation which, together with other anisotropy sources, could be able to align the filaments in a particular direction. In the case of the atomic target the only anisotropy present is the beam spatial profile, which explains the different experimenal multi-filaments orientation observed between the argon and the nitrogen targets. In order to follow the dynamic to have a better understanding of the difference between molecular and atomic behaviours observed, one could measure the multiple filamentation in different of gas mixture. The transition observable might be measurable and interesting since it would allowed an other degree of freedom (the ratio of species) to control the plasma effect on the dynamic.

Now that we can maintain supra-mJ single filament, we address the question of how the ellipticity degree influence the amount of initial peak power usable and how the nature of the target will limit or enhance this control? To understand this influence we performed a systematic measurement of the initial peak power to maintain one filament with respect to the ellipticity degree. The criteria we choose for the definition of single filament are: a single transverse mode, within this mode a supercontinuum generation of 300 nm with the same shape and a maintained longitudinal filament effective length in the visible. We observe systematically a visible length of 27 cm that corresponds to an effective filament length of at least two self-focusing cycles [39]. To estimate our initial peak power we measured the input pulse duration and the input energy. As it is shown in Fig. 4(a) the peak power could be increased with the ellipticity degree for both targets types in the same way maintaining one filament. It is also obvious, regarding the results, that each type of target cover a range of "allowed" input peak power. The different input power between the atomic and the molecular targets is again related with the ionisation process. It is known that for the case of random oriented nitrogen molecules and linear laser polarisation, the molecular-ADK theory [38] shows that the tunnel ionisation rate is 3 times lower than the argon case, so that the plasma effect is reduced in nitrogen. The input peak power can therefore be higher in the molecular than in the atomic target to obtain the same dynamics, as have been shown in our experimental data. For our purpose, the nitrogen target is the best choice to obtained a multi-mJ sub-10 fs laser source.

All the current filamentation post compression stage usually deals with linear input polarisation. To highlight what will be gained for these operated systems by converting their standard linear polarisation to a circular one we measure the ratio between input peak power necessary to maintain one filament in all the possible input ellipticity over the peak power in linear polarisation. The results are presented in Fig. 4(b) that shows a suprising effect with regard to the Fig. 4(a). Actually the maximum gain obtained in circular polarisation corresponds to a factor 2 in argon whereas in nitrogen it is limited to 1.6.

Note that the peak power measured here corresponds to the input peak power for generating one single filament. therefore It includes not only the Kerr effect but also the ionisation effect through the plasma formation. If now we consider only the Kerr effect, we found out that the self-focusing process is independent on the target nature (see Analysis section) with constant critical power gain of 3/2 for the circularly polarised laser. We conclude that the different gain obtained for each target is only due to ionisation that provides a total peak power gain superior to 3/2. (see Analysis section) with constant critical power gain of 3/2 for the circularly polarised laser. Therefore, the different gain obtained for each target is only due to jour set to 3/2.

Therefore, for the {argon, nitrogen} couple and for operating filament post-compression system, the maximum gain achievable by controlling the input polarisation state is obtained in



Fig. 4. (a: left) Input peak power for generating single filament as a function of the input ellipticity degree and (b: right) input power gain ellipticity dependence $\frac{P_m^{\varepsilon}}{P_m^{\varepsilon=0}}$ for argon (blue line) and nitrogen (red line). The blue dot line (respectively the red line) corresponds to the argon (respectively the nitrogen) target respons

the atomic target. This implies that depending on the laser peak power range available from a CPA system and the gain one would obtain, it is possible controlling the ellipticity and choosing the right target nature to achieve relevant output energy for applications. For multi-mJ fewcycles pulse our study shows that a circular input polarisation is needed together with a gas target of high ionisation potential and low ionisation rate. In that way we expect to increase the gain by increasing the output energy in a single filament mode towards multi-mJ range.

2. Analysis of the atomic and molecular Kerr effect for an elliptical field

It is well-known that for the case of a pure Kerr medium the critical power for collapse is $\frac{3}{2}$ times larger for a circularly polarised beam than for a linearly polarised one [40]. In this paper we study the filamentation dynamics while changing the polarisation of the beam. We have used atomic (argon) and molecular (nitrogen) media observing some differences between them. In this section we demonstrate that the origin of these differences cannot be related to the dependence of the nonlinear collapse dynamics with the polarisation because it is identical for atoms and molecules, although the Kerr effect is not the same for such media. In order to obtained the dependence of the critical power with the polarisation equations for a circularly polarised beam [40], obtaining the dependence of the nonlinear coefficient, n_2 , with this particular polarisation. In our case we have generalised this result by calculating the equation for a general elliptically polarised beam, including not only the ellipticity ε , but the possible different phase between both components ϕ .

2.1. Atomic target

The equations for slowly varying amplitudes of a general two polarisation components beam propagating in the direction z is [41]:

$$i\frac{\partial E_x}{\partial z} = -\frac{1}{2k_c}\nabla_{\perp}^2 E_x + \frac{k''}{2}\frac{\partial^2 E_x}{\partial t^2} - k_c n_2 \left[\left(|E_x|^2 + \frac{2}{3}|E_y|^2 \right) E_x + \frac{1}{3}E_x^* E_y^2 \right] \\ i\frac{\partial E_y}{\partial z} = -\frac{1}{2k_c}\nabla_{\perp}^2 E_y + \frac{k''}{2}\frac{\partial^2 E_y}{\partial t^2} - k_c n_2 \left[\left(\frac{2}{3}|E_x|^2 + |E_y|^2 \right) E_y + \frac{1}{3}E_y^* E_x^2 \right]$$
(1)

From this equation one can calculate the equations for the elliptically polarised components:

$$E_{+} = \frac{\varepsilon E_{x} - e^{-i\phi}E_{y}}{\sqrt{1 + \varepsilon^{2}}} \quad E_{-} = \frac{E_{x} + \varepsilon e^{-i\phi}E_{y}}{\sqrt{1 + \varepsilon^{2}}}$$
(2)

Assuming that we have a pure elliptically polarised beam, that is, that $E_{-} = 0$ for example, the equation for the other component has the following form:

$$i\frac{\partial E_{+}}{\partial z} = -\frac{1}{2k_{c}}\nabla_{\perp}^{2}E_{+} + \frac{k''}{2}\frac{\partial^{2}E_{+}}{\partial t^{2}} - k_{c}n_{2}\frac{\varepsilon^{4} + 4\varepsilon^{2}/3 + 1 + 2\varepsilon^{2}\cos\left(2\phi\right)/3}{\left(1 + \varepsilon^{2}\right)^{2}} \mid E_{+} \mid^{2}E_{+}$$
(3)

which indicates that the nonlinearity, and therefore the critical power for collapse, changes with the ellipticity state of the beam. In the limit of $\phi = \pi/2$ and $\varepsilon = 1$, circularly polarised beam, the factor converges to the well-known $\frac{3}{2}$. The same equation would be obtained for the other component. In summary, we have obtained the complete dependence of the critical power with the polarisation characteristics of the beam:

$$\frac{P_{CR}(\varepsilon,\phi)}{P_{CR}(0,0)} = \frac{\left(1+\varepsilon^2\right)^2}{\varepsilon^4 + 4\varepsilon^2/3 + 1 + 2\varepsilon^2\cos\left(2\phi\right)/3} \tag{4}$$

2.2. Molecular target

In the case of a molecular beam one has to take into account the retarded behaviour of the Kerr effect that could appear in the equation representing possible Raman processes. In this case, the equations for slowly varying amplitudes of a general two polarisation components beam are different:

$$i\frac{\partial E_x}{\partial z} = -\frac{1}{2k_c}\nabla_{\perp}^2 E_x + \frac{k''}{2}\frac{\partial^2 E_x}{\partial t^2} - k_c n_2 \left[\left(\int |E_x|^2 + \frac{1}{3}\int |E_y|^2 \right) E_x + \frac{1}{3} \left(\int E_x^* E_y + \int E_x E_y^* \right) E_y \right] \\ i\frac{\partial E_y}{\partial z} = -\frac{1}{2k_c}\nabla_{\perp}^2 E_y + \frac{k''}{2}\frac{\partial^2 E_y}{\partial t^2} - k_c n_2 \left[\left(\frac{1}{3}\int |E_x|^2 + \int |E_y|^2 \right) E_y + \frac{1}{3} \left(\int E_x^* E_y + \int E_x E_y^* \right) E_x \right]$$
(5)

where the expression $\int |E_i|^2$, or similar terms, indicate in a compact form the possible nonlinear retarded behaviour that the molecule could have, which expressed in a more extended way corresponds to $\int R(t') |E_i(t-t')|^2 dt'$ with $R(t) = (1-x_d)\delta(t) + x_d \int exp(-\frac{t}{\tau_d})/\tau_d$, where x_d indicates the contribution of the retarded Kerr effect in the medium and τ_d is the characteristic retarded time of the medium.

The surprising result is that calculating the equation for the same elliptically polarised components than before we have obtained exactly the same result than for the atoms, although the couple-mode equations for the two polarisation components are different for each case. In conclusion, the dependence of the critical power for collapse with the polarisation do not depend on the type of target.

Other formalism have been proposed by A. Couairon and co-workers in [42] to studied the filament formation of twin laser pulses in air. The system of coupled equations that they used differ from Eq. (5) in the cross phase modulation and the coupling term. We have check that

starting form the system of equations used by Couairon and co-workers and doing the same procedure shown above we obtain the same critical power for the general elliptical polarised components. Therefore, even with different descriptions for the cross phase modulation term and of the coupling between the two polarisations we conclude that the dependence of the critical powre with the polarisation is the same for atoms and for molecules. We really believe that a further discussion on the correct description of the cross phase modulation and the coupling between the two polarisation must be done

3. Conclusion

We managed to define and find experimental conditions to produce supercontinuum of 300 nm consistent with few-cycles pulses generation through filamentation in gas with a supra-mJ output energy. This is done controlling the ellipticity degree of the input laser pulse and choosing a gas target that can afford the available laser input peak power. We demonstrate the capability of polarisation control to enhance the limitation of the input power by a factor 2 maintaining one single filamentation propagation mode and we highlight the importance of choosing the right target with respect to the range of the input peak power available. We also discuss the implication of circular polarisation on the induced filamentation and we highlight that ionisation is responsible for the different behavior attained between atomic and molecular targets. Besides, to build high power few-cycles pulse sources by filamention, the polarisation controlled filamentation technique is robust and easy to implement on any standard CPA system. It permits to break for the first time the mJ output energy limit for supercontinuum generation maintaining one single filament. Therefore we think that our technique is adapted to high power few-cycles pulses generation.

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