Some Consequences during the Propagation of an Intense Femtosecond Laser Pulse in Transparent Optical Media: a Strongly Deformed White-Light Laser

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Abstract—We present some results concerning the propagation of intense femtosecond Ti:sapphire laser pulses in matter and especially in air. We show that the supercontinuum created during the propagation preserves the spectral coherence of the initial pulse and both together form a chirped-white-light laser. Furthermore, we detected the photoemission of N$_2$ and N$_2^+$ to measure the intensity dependence of a focused laser pulse in air along the propagation distance and we observed a multiple refocusing. Finally, we underlined a clear charge separation in the plasma created a focused pulse in air giving rise to a short electric pulse.

INTRODUCTION

In recent years, the phenomenon of the propagation of strong ultrashort laser pulses in a transparent optical medium and especially in the atmosphere has been studied both experimentally and theoretically [1–5]. As a result, a qualitative picture has emerged which is able to explain the fundamental physical mechanisms of the universal phenomenon [2–4] that leads to self-focusing, filamentation, supercontinuum generation, conical emission and plasma generation. Each effect has been extensively studied to a better understanding of the phenomenon and in this paper, we contribute to that by studying some properties occurring during the propagation of intense ultrashort laser pulses in transparent optical media. Especially, we will focus on the nature of the supercontinuum source by studying the coherence properties of the white-light generated by self-phase modulation in liquids and we will show that the supercontinuum source is a spectrally pure coherent source. Then, we will present some results reflecting the behavior of the beam when the intense femtosecond pulses are focused in air, i.e., a multiple refocusing of the beam. Finally, we will show how the charges created by multiphoton ionization by intense focused laser beam in air are separated leading to the existence of an ultrafast electric field in the column of plasma. We will then conclude by showing how the intense short laser pulse propagating in matter is strongly deformed in space and time but still remains a laser.

THE LASER SYSTEM

All the experiments that we present have been performed by an ultrafast laser system and a schematic is shown in Fig. 1. It is based on a Kerr-lens-mode-locked Ti:sapphire oscillator (Clark MXR NJA-4) pumped by a cw all-solid-state-diode-pumped frequency-doubled Nd:YVO$_4$ laser (Coherent Verdi). The output pulses are amplified by chirped pulse amplification in a regenerative amplifier pumped by a 1 kHz Q-switched Nd:YAG laser. The beam is then sliced into two beams. One of them is sent directly to an optical compressor; the compressed pulses centered at $\lambda_0 = 800$ nm are 170 fs long (FWHM) with a time-bandwidth product $\Delta \nu \Delta t = 0.57$ and the energy is more than 1 mJ/pulse. The other beam is amplified in two stages. A two-passes Ti:sapphire preamplifier pumped by a 10 Hz Nd:YAG laser (Continuum Surelite) amplifies the pulses up to 15 mJ. A four-passes Ti:sapphire amplifier is then used to increase the pulse energy to more than 160 mJ. Pulses are then recompressed by a grating optical compressor down to 220 fs.

SPECTRAL COHERENCE PROPERTIES OF THE SUPERCONTINUUM

White light generation in transparent optical media is one of the most popular methods of broadening the
spectrum of laser sources. Briefly, the spectrum is extended by self-phase modulation from the near UV to the near infrared in such a way that this source is called a supercontinuum [6, 7] and in the case of ultrashort laser pulse, this supercontinuum is accompanied by a filament. This white light supercontinuum has been used as a seed for further amplification in multipass dye amplifier [8] or OPA’s [9], as a tunable source for surface second harmonic generation [10], for resonant pump-probe experiment in transient absorption spectroscopy [7] or as broad source for ultrashort pulse generation down to less than 10 fs [11]. Despite this extensive use of the white light supercontinuum, researchers do not see the need to verify that such a source is effectively a coherent source. Because the supercontinuum is generated by a coherent laser source and involves instantaneous nonlinear effects, it is tacitly accepted that it is also a coherent source and the applications are the best proof. In this section, we show that in terms of spectral coherence, the supercontinuum is indeed a coherent source.

To achieve that we use the technique of the spectral interferometry to measure the spectral coherence of the supercontinuum. When two beams at the output of a Michelson interferometer are sent into a spectrometer, the superposed spectral components interfere on the square-law photodetector and give rise to a spectral modulation. This modulation depends on the delay between the two beams and is observable even when the path difference is longer than the temporal coherence length. To describe this experiment, the concept of the spectral visibility and the spectral correlation introduced by Mandel and Wolf [12] is used and more details on the development can be found in [13]. Briefly, when the two beams interfere in a spectrometer, the spectral intensity distribution $I_v (r)$ is given by

$$I_v (r) = I_v^1 (r) + I_v^2 (r) + 2 \sqrt{I_v^1 (r) I_v^2 (r)}$$

$$\times |\mu| \text{sinc} (\pi \Delta \nu \tau_{12}) \cos \left[ \alpha + \beta - 2 \pi (v + \Delta \nu \frac{\tau_{12}}{2}) \right].$$

where $I_v^1 (r)$ and $I_v^2 (r)$ are the spectral intensity of each field, $\mu$ the complex degree of spectral coherence with a phase factor $\beta$, $\alpha$ a phase factor, $\tau_{12}$ the time corresponding to the path difference between the beams and $\Delta \nu$ the resolution of the spectrometer. The visibility of the spectral modulation is then

$$V (r, v) = |\mu| \text{sinc} (\pi \Delta \nu \tau_{12}).$$

Thereby, the spectral modulation is detected on a finite interval $\tau_{12}$ until a threshold where the spectral modulation cannot be distinguished from the noise fluctuations. This defines the experimental threshold visibility $t_v$ and the interval of time where the visibility above the threshold is

$$\Delta \tau = \frac{2 \sqrt{6}}{\pi \Delta \nu |\mu|}. \quad (3)$$

For a low spectrally coherent source, the ratio $t_v / |\mu|$ will be larger than in the case of a highly spectral coherence source. Hence, the width of the visibility $\Delta \tau$ will reflect the differences between the spectral coherences of different sources as long as the experiment is performed under identical conditions. By analogy with the temporal coherence time, we call the finite interval of time where the spectral modulation is visible the spectral coherence time (and by extension the spectral coherence length).

Because of the universality of white light supercontinuum generation, we choose to work with liquid which is easier to handle. The 1 KHz beam is focused by a 10-cm lens through a fused-silica window into a cell filled with a liquid (water, CCl$_4$ or methanol). The laser input energy is adjusted by variable neutral density filters in such a way that only one filament can be seen. In water, it is adjusted to 3 mJ whereas in CCl$_4$ and methanol, only 0.2 mJ are used. The white output beam is then collimated by a 10 cm lens and sent to the Michelson interferometer followed by a grating spectrometer. The spectral coherence lengths are measured by using the spectral interferences between two beams at the output of a Michelson interferometer (Gaertner 458-N) sent collimated onto the entrance slit (70 μm wide) of a grating spectrometer (SPEX 1870 with 1200 grooves/mm. A photomultiplier tube detects the spectrum and the rotation of the grating scans the spectrum across the white-light spectral range (350–900 nm). The spectrum is then recorded across a width of 1 nm around a central wavelength and the delay is changed as long as the spectral modulation appears. This allows us to determine the spectral coherence time. In Fig. 2, we plot the spectral coherence length as a function of the central wavelength measured for the three supercontinuum sources. In all the cases, we observe that the results are identical. In the range from 400 to 800 nm, the spectral coherence length increases
linearly with the wavelength and this can be explained as follows. The spectral coherence length is inversely proportional to the spectral resolution $\Delta \nu$ of the device which depends on the number of illuminated grooves on the grating. It emerges that the spectral coherence length varies linearly with the central wavelength. This is experimentally observed (Fig. 2) and the slope depends on the ratio between the threshold level $t_0$ and the degree of spectral coherence $|\mu|$. At 900 nm, the spectral coherence length decreases. This is because at this wavelength the spectral intensity drops very quickly to the noise level in our experiment. This makes $t_0$ larger, hence a larger ratio $t_0/|\mu|$ and thus a lower value of the spectral coherence length. In the linear range, the fluctuations of the spectral intensity being roughly constant $t_0$ is roughly constant. Hence the modulus of the complex degree of spectral coherence $\mu$ is constant for every frequency component present in the spectrum. According to Mandel and Wolf [12], this means that the source is cross-spectrally pure and in this case the modulus of the complex degree of spectral coherence is equal to the modulus of the complex degree of temporal coherence. As we mentioned above, the results are universal for the three supercontinuum sources; the nonlinear effects involved in the white-light generation are the same in any liquid and the coherence properties of the supercontinuum are not affected by the medium where it is created [14]. Moreover, we did the same experiment under the same conditions with the initial Ti:sapphire laser pulses at 800 nm and we plot the result on the same graph. As we can see, the spectral coherence length of the supercontinuum at 800 nm and that of the laser are the same. We conclude that the spectral coherence of the initial laser and the supercontinuum are the same, i.e., the spectral coherence of the initial laser pulse is preserved over the whole spectrum by the processes creating the supercontinuum. The supercontinuum is thus a coherent source and as long as the initial pulse is called a laser pulse, the supercontinuum should also be qualified as a laser.

**REFOCUSING OF FEMTOSECOND Ti:SAPPHIRE LASER BEAM DURING THE PROPAGATION IN AIR**

As mentioned above, a qualitative picture emerged to explain the fundamental mechanism involved in the propagation of intense short pulses in optical transparent media. However, more rigorous models predicted some new phenomena such as multiple refocusing which need more careful measurement of the intensity distribution in the filament for a quantitative comparison. Due to the high intensity in the filament, incorporating measurement systems directly into the beam is nearly impossible, therefore the intensity distribution should be determined from the measurement of some quantities outside the filament. In this section we offer the results of our measurement that reflects the gross intensity in the filament as a result of the propagation of focused femtosecond laser pulse in air. These results confirm the phenomenon of multiple refocusing of the laser pulses after the geometrical focus [5].

The laser pulse is focused through 1.5 m lens in air. The diameter of the input beam is varied using a diaphragm before a spatial filter followed by the compressor. The beam has a Gaussian profile and the diameter of the beam is defined at the $1/e^2$ level of the peak intensity. The filament which is created as a result of focusing the laser pulse is visible to the naked eyes in a range of 20 cm spanning the focal region. The radiation from the filament is analyzed spectrally. The central part of the filament is imaged onto the entrance slit of a spectrometer which is calibrated for the spectral response. A sample spectrum of the radiation is presented in Fig. 3. As it is observed, there is no detectable continuum radiation, characteristic of collisional ionization, in the spectrum. The observed violet degraded lines are assigned to two band systems; the second positive system of $N_2$ ($C^3\Pi_u - B^3\Pi_g$ transition) and in the first negative band system of $N_2^+$ ($B^2\Sigma_u^+ - X^2\Sigma_g^+$ transition) respectively. We believe the second positive band system results from the $N_2$ molecules excited through dynamic multiphoton absorption in the strong laser field followed by collisional population transfer to the $C^3\Pi_u$ state. The first negative band system is radiated...
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by $N_2$ molecules which are excited through an inelastic collision with the re-scattered electron. The electron is freed from $N_2$ molecule via multiphoton ionization (MPI; tunnel ionization is included in this general appellation) and as a result of the interaction with the laser field is accelerated back to its parent ion. Later in this section, we will discuss the significance of the signal detected, and it will be shown that it is a monotonic function of the intensity in that region of the filament beneath the detector.

The total intensity of the radiation from a restricted part of the filament (restricted by a slit, with an opening of 0.2 mm, situated at $R = 1$ cm above the filament) at a distance $z$ from the lens in a frequency range of $330 < \lambda < 460$ nm (filtered by a colored filter) is detected by a photomultiplier. The amplitude of the output of the detector is taken as the measured signal. In Fig. 4, the results of our measurement using two different beam sizes (11 and 4 mm) is presented. When using the 11-mm diameter beam (Fig. 4a) the peaks of the plots occur at a position before the geometrical focus indicating self-focusing. The distance of the focusing point from the geometrical focus increases with the energy of the pulse. This is consistent with the prediction of the moving focus model [15]. After focusing, the beam rapidly diverges as indicated by the decrease of the signal. If the energy of the pulse is sufficient, the beam divergence stops along some distance before it starts to diverge again. This is clearly seen from the first shoulder of the curves after the geometrical focus. At higher energies of the pulse, more than one shoulder could be observed. Thus, the detailed behavior of the beam strongly depends on the pulse energy. When using the 4-mm diameter beam (Fig. 4b), again the peak (self-focus) moves towards the lens, though with an amount more appreciable than the case of larger beam diameter. This is well anticipated noting that in this case, the input power is about 7.6 times (= the ratio of the cross sections of the two beams) higher than the case of a larger beam with the same energy and pulse duration. After self-focusing, a more interesting situation arises; the signal level increases clearly through a second peak indicating that the beam after a slight divergence focuses again. This is a clear demonstration of the phenomena of multiple refocusing [2–5]. In particular, the prediction of refocusing in [5] in the case of a focusing geometry is clearly observed here. By analogy, the shoulders of Fig. 4a might be considered as indicating the refocusing. If so, the appearance of multiple shoulders in the case of high-energy pulses could be considered as the occurrence of multiple refocusing. Thus, our measurement agrees qualitatively with the results of the simulations of [5]. However, a quantitative agreement is unlikely. At the high intensities present in the filament, higher order nonlinearities might become as important as the third order nonlinearity. For example $\chi^{(5)}$ could contribute to the defocusing and thereby diluting the self-focusing effects of $\chi^{(3)}$. In numerical simulations the effects of high order terms are neglected.

As was noticed by comparing the two Figs. 4a and 4b, for a given energy of the pulse the manifestations of the nonlinear propagation effects (early focusing, refocusing and slow divergence after focusing meaning filamentation or a streak of moving foci) is weaker in the case of the beam with larger diameter, or more precisely shorter Rayleigh range. This enables us to study the significance of the signal. We thus performed a separate experiment by increasing the diameter of the beam to 2 cm and focusing it by a 1-m focal lens in air. In this case due to the short Rayleigh range (=20 mm) as compared to the case of Fig. 4 the effect of self focusing before the geometrical focus is negligible, such that the distribution of intensity at the focal region does not deviate strongly from the one expected in vac-
uum. This fact is readily observed in the inset of Fig. 5, which represents the detector signal at a distance from the lens. Even at the appreciably large value of the pulse energy, 20 mJ, the curve is symmetric and focusing practically occurs at the geometrical focus. We measured the laser pulse energy dependence of the signal with the detector placed above the focal point (z = 100 cm). The result is shown as open circles in Fig. 5.

Then, in a vacuum chamber the intensity dependence of the total number of \( N_2^+ \) ions is measured (for the details of the experiment see [16]), and the result is shown as dots in Fig. 5. The photon signal has been shifted vertically such that the turning point coincides with the saturation point of the ion curve and horizontally by a factor of two towards higher pulse energy. The complete overlap of the two curves indicates two aspects. Firstly, the photon signal reflects the gross intensity in the filament and is a monotonically increasing function of the pulse energy. Secondly, the probability of photon emission scales in the same way as the probability of multiphoton ionization to \( N_2^+ \) molecular ion at a pulse energy twice lower. We can thus take advantage of this observation to propose the following for theorists to test the validity of their propagation model. We could say that the photon signal generated at a theoretically calculated intensity \( I(r, t) \) in the filament is proportional to the ion signal generated at intensity \( I(r, t)/2 \) in the vacuum chamber. We can calculate the ion number, \( N(I/2) \), in this case precisely by the following formula:

\[
N(I/2) = \int dV \left( 1 - e^{-\frac{-W_{\text{fit}}(r,t)/2}{W}} \right), \tag{4}
\]

where \( W \) is the rate of MPI of \( N_2 \) molecule. Recently, we have measured the rate of the MPI of this molecule and concluded that it is very well predicted by the PPT model [17] with \( Z_{\text{eff}} = 0.95 \) (\( Z_{\text{eff}} \) is a fitting parameter to simulate the effective Coulomb potential felt by the electron that tunnels out. This parameter depends principally on the random orientation of the ground state neutral molecule. For more details see [18]). As we have concluded earlier, \( N(I/2) \) thus obtained must be proportional to the signal we measure in Fig. 4. Thus, the calculated ion signal as a function of \( z \) should fit those curves in Fig. 4 if the model were correct.

In conclusion, we have measured the radiation emitted by the \( N_2 \) molecules and \( N_2^+ \) molecular ions interacting with the laser pulse in the filament. The result of the measurement gives the gross intensity in the filament as a function of the propagation distance showing a multiple refocusing which agrees qualitatively with the results of the simulations of [5].

Recently, some research groups [19–21] devised a simple method to measure the pulsed fields created as a result of the interaction of a focused laser pulse in air. In their studies, the emphasis was upon the interaction of long laser pulses (\( \mu s \), ns and 700 ps), where the ionization of the molecules and generation of plasma is due to the acceleration of electrons through inverse bremsstrahlung followed by inelastic collisions with molecules. When the density of the carriers in the interaction region exceeds a threshold value [22] there will be avalanche ionization which creates a high density plasma. The conclusion of [19–21] is that the plasma created by this ionization process is not locally neutral and therefore the interaction volume possesses a dipole or quadrupole moment. By placing an antenna in the vicinity of the interaction volume, they were able to measure the field created by this charge distribution.

Naturally the next step would be to extend these types of studies to the case of a plasma created through the interaction of short laser pulses with air because the ionization mechanism is different. It is thus expected that the produced plasma will have characteristics different from the plasma created by long laser pulses. It is known that the characteristic time for the collisions in air is \( \sim 1 \) ps [22], so in the case of pulses with duration around 100 fs, the collisional ionization followed by
Avalanche will not have an appreciable contribution in the generation of the plasma and it will be dominated by multiphoton ionization (MPI) of the molecules. These characteristics enable us to determine the origin of multiple electric moments, a task which in the case of long laser pulses has not been established [19–21].

Motivated with this expectation, we measured the pulsed electric field produced in the vicinity of the plasma column which is created during the propagation of a focused 220 fs Ti:Sapphire laser pulse in air. Our measurement indicates that the plasma column possesses a dipole moment. This might be explained by the theoretical findings of Goreslavsky and Narozhny [23]. According to their calculation, the electrons created inside the short pulse through the MPI will be accelerated forward resulting in an effective separation of electrons and positive molecular ions.

Experimentally the plasma column is created in air by focusing the 220-fs Ti:Sapphire laser pulse through a 1.5-m lens and is faintly visible to the naked eyes in a range of about 20 cm spanning the focal region. We place the dielectric shielded inner core of a 50 Ω coaxial cable at a distance of 0.5 cm from the plasma column and the signal detected by this antenna is sent to a 1 GHz oscilloscope having a 50 Ω input impedance (Tektronix 7834). As shown in Fig. 6, the duration of the signal is less than 1 ns (which is the detection limit of our oscilloscope). The current experiment clearly shows that the signal is electric in nature.

By measuring the amplitude of the signal as a function of distance from the geometrical focus of the lens, the dependence of the signal on the propagation distance can be determined. Figure 7 shows the results for the case of 36-mJ pulses focused by a 100-cm focal length lens (Fig. 7a) and by a 150-cm focal length lens (Fig. 7b). Because the distance between the wire detector and the plasma column is small, we conclude that the detected field distribution shown in Fig. 7 corresponds in fact to the net charge distribution in the plasma column. In other words, if a positive net charge is suddenly created in the region near the wire, it will induce a current in the wire which will give a positive signal S(t). If a negative net charge is created near the wire, then the induced current will be in the opposite direction and the signal will be negative. This experimental measurement of the net charge distribution is the first step to make in order to fully reconstruct the positive and negative charge distributions in the plasma column. The next step consists of measuring the positive ions distribution. The knowledge on the distribution of the positive ions in this column could then allow us to fully reconstruct the electron distribution.

In the same manner as we presented in the previous section, we measured the distribution of the N₂⁺ ions by measuring the photoemission coming from the plasma column (for more detail, see [24]) where the strength of the photoemission signal integrated over all emission frequencies is proportional to the number of the created N₂⁺ ions. From the distribution of the positive ions and the net charge (Fig. 7) we can calculate the electron distribution in the plasma column by subtracting the net charge distribution from the N₂⁺ ions distribution. As discussed above, the entire positive ion distribution is proportional to that of N₂⁺. This allowed us to scale the net charge distribution in such a way that the calculated number of electrons equals the number of positive ions. The resulting curves shown in Fig. 8 demonstrate that, in the plasma column, the positive charges and the electrons are separated in the longitudinal direction. The mass of the positive charges being much larger than...
that of the electrons, our observation implies that it is the electrons that have been displaced through their interaction with the laser pulse. This phenomenon might be explained using the theoretical findings of Goreslavsky and Narozhny [23]. After the MPI of the molecule in the leading front of the pulse, the electron is drifted along the propagation axis in the forward direction because of ponderomotive acceleration. When it is in the back-end of the pulse, it decelerates until it stops when the pulse has entirely passed it. As a result, a net charge separation is established, creating a large dipole in the plasma column. As can be seen in Fig. 8, the phenomenon of charge separation is present in both cases independently of the focusing conditions, which implies the universality of the charge separation phenomenon. After the laser pulse has passed, the strong electrostatic restoring forces bring back together the separated charges, resulting in an oscillatory dynamics of the electrons that will cause them to radiate. This radiation corresponds to an electromagnetic pulse faster than 1 GHz.

CONCLUSION

In this paper, we analyzed the propagation of an intense ultrashort laser pulse in liquids and in air. We showed that the spectral coherence of an initial laser pulse is preserved in the supercontinuum created by self-phase modulation in matter. This phenomenon is universal, independent of the medium, and it has been shown that the supercontinuum in air is composed of chirped spectral components [25]. Our recent analysis [26] shows that the supercontinuum from any transparent optical media including gases [14] is a result of the severe spatio-temporal deformation of the pump pulse undergoing self-focusing and interacting with the self-generated plasma [2–4]. Furthermore, we measured the photoemission coming from the plasma column created by multiphoton ionization and by varying the position along the filament, we showed a multiple refocusing of the beam. This strengthens the idea that the beam is indeed spatially very strongly deformed during its propagation. In addition, the light beam creates a separation of the ions and electrons in air and the laser-induced dipole moment will oscillate that give rise to a short electric pulse with a duration less than 1 ns. In conclusion, the laser pulse propagating in optical transparent media become a chirped white-light laser self-refocusing several times and inducing the charges left behind to emit a short electric pulse.

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