Simple 3-D characterization of ultrashort laser pulses

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Abstract: The space and time distribution of electromagnetic energy is essential information for any laser pulse applications that require precision. Although many instruments quantify the temporal profile of ultrashort laser pulses, they are generally limited to space-averaged measurement. In this work, we present an extremely simple technique to characterize the spatial distribution of fluence, pulse duration and chirp of ultrashort light pulses. This technique is based upon imaging the two-photon fluorescence distribution generated by the laser pulse as it propagates through a dispersive medium. It is expected that this technique will provide to less specialized users a precise in-situ analysis of their ultrashort laser beam.

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1. Introduction

Temporal and spatial distribution of ultrashort laser pulse energy is crucial information for applications such as the investigation of electrons dynamics [1], the coherent control of chemical reaction [2-4] or the phototherapy with femtosecond (10⁻¹⁵ s.) laser pulses [5]. The three most important parameters for a laser pulse are the spatial distribution of the laser fluence (energy per unit area), the pulse duration and the chirp (the frequency change across the pulse). Although recent advances in ultrashort pulse measurement [6-8], present techniques are still complex and limited to a narrow range of wavelengths. These techniques based upon the correlation of ultrashort laser pulses [7-9] measure the spectrum of the generated second harmonic and through a complex algorithm, the envelop amplitude and the phase of the electric field can be retrieved. The precision provided by these techniques is difficult to achieve with other methods, but these correlation measurements are often averaged across the section of the laser beam. Thus, to increase the reliability and the precision of experiments using ultrashort laser pulses, such informations on the spatial and temporal distribution of the laser pulse are therefore the major challenge [10, 11].

2. Experimental technique

In the present work, an efficient and extremely simple approach is presented to characterize precisely the space-time distribution of a laser pulse. The basic idea is very simple: when a short optical pulse (necessarily composed of many frequencies) propagates through a dispersive medium, the group velocity dispersion changes the pulse duration. This change depends on the initial pulse duration and chirp. Therefore, the intensity of the laser pulse (fluence/pulse duration) will also be changed and this can be measured by any kind of non-linear excitation processes. An example is the two-photon fluorescence of dye molecules dissolved in dispersive medium such as water or methanol (see Fig. 1(a)). In our simulation, we consider a Gaussian pulse envelop for which the expression of the electric field is of the form

\[ \psi(x,y,t) = \psi_0(x,y) \exp\left( -\frac{2}{\sigma^2} \right) \exp\left( i\omega t - k z \right) \]

where \( \sigma \) is the two-photon absorption cross-section of the dye, \( \alpha \) and \( k \) are the one-photon absorption and the second order of dispersion of the solvent medium, respectively. Because these material parameters are known (or measurable) we are able to derive the spatial distribution of the laser fluence \( S(x,y) \), the initial pulse duration \( \tau_0(x,y) \) and the chirp \( \chi_0(x,y) \) with a straightforward fit-routine. The x-y plane is transverse to the propagation axis z. This analysis can easily be generalized to any arbitrary temporal profile [12]. For pulse duration from 1ps down to few-cycle and over a propagation distance of 3cm in methanol or water, we verified numerically that the higher-order dispersion terms were negligible and induced deviation less than 5% from the Eq. (1) for the time-integrated fluorescence signal. Using this technique we have a tool for the spatial and temporal beam characterization applicable for pulse lengths of a few femtoseconds (fs) up to about a picosecond.
accessible wavelength range depends on the two-photon absorption spectra of the fluorescing materials. For dye molecules the wavelength spans from the visible up to the near-infrared. Maximum laser intensities should be below 100 GW/cm$^2$ in order to avoid saturation and nonlinear propagation effects [13]. Then, to retrieve the three dimensional distribution of the laser energy (the third dimension being the pulse duration multiplied by the speed of light), the slit in Fig. 1(a) is scanned across the beam profile and for each position of the slit the fluorescence distribution along the propagation axis can be fitted with the Eq. (1).

![Diagram of measurement setup](image)

**Fig. 1.** (a) Illustration of the measurement setup. The fluorescence induced by the femtosecond laser pulse propagating in dye solution is imaged by the CCD camera. The slit is used to characterize individually each slice of the beam profile. The dimensions of the slit are adjusted such that the diffraction is negligible. (b)-(d) Simulated two-photon fluorescence distribution induces by a laser pulse of 7 fs at full width at half maximum, centered at 737 nm and propagating in methanol/dye solution. (b) The initial chirp is positive ($\zeta_0 = +3$), that is, with the longer wavelengths on the leading edge of the pulse and the shorter ones on the trailing edge. (c) The laser pulse is transform limited ($\zeta_0 = 0$). (d) The initial chirp is negative ($\zeta_0 = -3$), that is, with the shorter wavelengths on the leading edge of the pulse and the longer ones on the trailing edge.

The development of the two-photon fluorescence signal along the propagation axis $z$ is directly related to the initial characteristics of the pulse. The longitudinal distribution of the fluorescence decreases monotonically if the initial chirp is positive (see Fig. 1(b)). For negatively chirped laser pulses the fluorescence signal first increases and reaches a maximum at the position (see Fig. 1(d)) [14]

$$z = \frac{-\zeta_0}{1 + \zeta_0^2} \left( \frac{t_p^2}{k_z^2 4 \ln 2} \right)$$

from whereon it decreases according to the transform-limited pulse duration (see Fig. 1(c)). The shorter is the initial pulse duration, the more pronounced is the fluorescence decay. In fact an eye inspection of the fluorescence pattern already provides a first estimate of the laser pulse parameters. In case of composite laser pulses we have to use a sum of Gaussian pulses with different initial parameters (fluence, pulse duration and chirp) for fitting the fluorescence signal.
3. Results and discussion

First, to verify the reliability and the precision of the technique presented in this work, we performed an experiment using a Ti:sapphire chirped pulse amplification laser system, which generates femtosecond laser pulses with a central wavelength at 807 nm and a spectral width of 28 nm (see Fig. 2(a)), a repetition rate of 1 kHz, an energy per pulse of 50 μJ and a transform limited pulse duration of 48±5 fs (measured with a scanning autocorrelator) at full width at half maximum (FWHM). We used typical laser intensity below 100 GW/cm² in order to avoid the intensity saturation of the dye and the propagation length in the dye solution was much shorter than the diffraction length of the laser beam.

The longitudinal distribution of the fluorescence averaged across the section of the laser beam is shown in Fig. 2(b). The red line in Fig. 2(b) corresponds to the best fit according to Eq. (1). From the measurement of the fluorescence, we retrieved a pulse duration of 46.7 fs and the laser pulse was slightly negatively chirped by ζ₀ = -0.03. Clearly this is in excellent agreement with the autocorrelation measurement, yielding 48±5 fs. Figure 2(c) and 2(d) show the sensitivity of the fitting (data from Fig. 2(b)) by slightly varying the initial pulse duration (t₀) and the chirp (ζ₀) during the fitting process. We can see from the Figure 2(c) and 2(d) that this technique yields reliable data on pulse duration and chirp. The error for the inferred pulse duration is less than 2% by assuming a Gaussian temporal profile.

Second, to exemplify more our method we present the experimental results and the accompanying analysis for a few-cycle laser pulse. The laser system used consists of three amplification stages and it is seeded by the pulses from a Ti:sapphire oscillator. The 2mJ pulses after the final amplification stage are compressed to 12 fs in a prism compressor. In order to generate few-cycle pulses, the 12fs pulses are focused in a hollow-core fibre. The fibre was filled with Neon at a pressure of 1.5 bar. The length of the fibre is 1m and the core diameter is 0.3mm. Pulses at the output of the fibre are temporally recompressed to 7±1fs (measured by an interferometric autocorrelator) at FWHM by means of ultrabroadband
chirped mirrors. The energy per pulse used for the current experiment was 30 μJ and the peak intensity was 20 GW/cm². The spectral characteristics and the second-order autocorrelation of the laser pulse are shown in Fig. 3. The autocorrelation reveals that the few-cycle pulse is superimposed on a long pedestal pulse, but this is only profile-averaged information. The side fluorescence image induced by this laser pulse is shown in Fig. 4(a). Figure 4(b) shows the longitudinal distribution of the fluorescence (black line) taken at the center of the laser beam. In our analysis, we used three independent Gaussian laser pulses to simulate the recorded fluorescence trace within a maximum deviation of 1.3%. The use of a fourth independent pulse for the fitting was meaningless because it contained less than 1% of the total laser energy which was below the precision limit for the energy/fluence measurement. From the fitting curves in Fig. 4(b), we retrieve a negatively chirped pulse (red line) of 7.2 fs and ζ₀ = -2.6. We find also a second laser pulse (green line) of 30 fs and ζ₀ = -0.11 that contains 40% of the total energy. Finally, there is a third laser pulse (blue line) of 51 fs and ζ₀ = -1.02. This pedestal pulse contains about 10% of the total laser energy. We note here that only 50% of the total energy appears in the few-cycle laser pulses and this observation is supported by the autocorrelation trace shown in Fig. 3(b).

![Fig. 3. (a) Spectral distribution of the laser pulse. (b) Autocorrelation signal. The retrieved pulse duration from (b) is 7±1 fs at full width at half maximum. We note from the autocorrelation trace that there is a pedestal pulse of unknown duration that is superposed with the short laser pulse. arb. unit, arbitrary unit.](image)

Furthermore, the fluorescence pattern in Fig. 4(a) is slightly curved and tilted along the transverse direction. This suggests that the pulse duration and the chirp are not uniform across the horizontal profile. A two-dimensional fit according to Eq. (1) allows to reconstruct the horizontal variations of the pulse parameters for both the few-cycle and the pedestal laser pulses. The results are displayed in Fig. 4(c). We can clearly see that the chirp and the pulse duration are not uniform across the horizontal direction although both pulse fluences nearly resemble to smooth Gaussian beam modes. For the few-cycle laser pulse, the pulse duration varies from 5.6 fs to 9.5 fs along the horizontal profile, which corresponds to a relative deviation of ±25% from the profile-averaged pulse duration. From the spatially integrated fluorescence distribution, the effective pulse duration is 7.4fs at FWHM, which corresponds to the inferred 7±1fs pulse duration measured with the interferometric autocorrelator.

We note also in Fig. 4 that more than 80% of the fluorescence at the onset of propagation (0-2 mm) is generated by the short pulse (~7fs). While at the end of the dye cell, more than 95% of the fluorescence is generated by the pedestal pulse (~50fs). Thus, by imaging the transverse section of the fluorescence distribution at the beginning and at the end of the dye cell, we could also image directly and individually the beam profile of the short and the pedestal pulses.
Fig. 4. Characterization of a few-cycle laser pulse. (a) False color representation of two-photon fluorescence induced by a laser pulse (7±1 fs measured with an autocorrelator and the central wavelength is 737 nm) propagating in a solution of Fluorescein 27 dissolved in methanol (concentration 2.4 x 10^17 molecules/cm^3). The measured (black line) longitudinal fluorescence distribution at the center of the beam is shown in (b). The purple curve (Sum of fits) is the best fit. The red, green and deep blue curves in (b) correspond to the fluorescence induced by individual pulses. The transverse distribution of pulse duration, chirp and laser fluence for the few-cycle and pedestal laser pulses are shown in (c).

Another advantage of this technique is that it can be easily applied to any laser pulse wavelengths ranging from the visible to the infrared. The longitudinal distribution of the fluorescence (black line) taken at the center of the laser beam for a visible, near-infrared and infrared laser pulses are shown in Fig. 5(a) to 5(c), respectively. The corresponding spectra are shown in Fig. 5(d) to 5(f) on the right-hand side of the longitudinal fluorescence. In Figure 5(c), the one-photon absorption (0.46 cm^-1) around 1.32 μm from the solvent used for the dye solution was taken into account for the decrease of the laser intensity. For each measurement done at different wavelengths, a precise measurement of the pulse duration and chirp was obtained with this technique. For these measurements, we needed only one independent Gaussian laser pulse to simulate the recorded fluorescence trace within a maximum deviation of few percents. The error on the retrieved pulse duration was inferior to 1 fs for the results shown in Fig. 5.
Fig. 5. The longitudinal two-photon fluorescence distribution (black line) of the laser beam is shown in (a) for a laser pulse centered at 560 nm and generated through four-wave mixing in air. The dye solution was Coumarin 440 dissolved in methanol at 4x10^{-4} mol/L; (b) for the output pulse from a Ti:sapphire oscillator centered at 800 nm. The dye solution was Fluorescein 27 dissolved in methanol at 4x10^{-4} mol/L; (c) for an infrared pulse centered at 1.32 μm and generated from an optical parametric amplifier. The dye solution was Nile Blue dissolved in methanol at 10^{-3} mol/L. The red curves are the best fits and the parameters are indicated on the graphs. The normalized spectral intensity distribution of the laser pulse are shown in (d) for the four-wave mixing pulse, (e) for the Ti:sapphire laser pulse and (f) for the optical parametric laser pulse. arb.unit, arbitrary unit.

4. Conclusions

Finally, this new and simple technique consists of a dispersive medium containing two-photon absorbing dye molecules acting as the spectator of the local pulse intensity and an imaging system to measure the emitted fluorescence. The information on the spatial distribution of the chirp, pulse duration and fluence measured with our method were not accessible with previous correlation techniques. In fact this new and simple method can be use for a wide range of pulse parameters even in the case where the laser pulse energy is only few nanojoules and for wavelengths ranging from the visible to the infrared. However, this technique has limitations because it measures the time-integrated fluorescence signal. In particular, it can not resolve the time delay between independent pulses and the pulse front tilt. Nevertheless it can be used as a complementary and simple screening technique to measure the spatio-temporal distribution of ultrashort laser pulse. This new technique will certainly results in a better characterization of femtosecond laser beam and pushes forward the precision limit for ultrashort laser pulses applications.

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