Three dimensional imaging of short pulses

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Abstract

We exploit a slightly non-collinear second-harmonic cross-correlation scheme to map the 3D space–time intensity distribution of an unknown complex-shaped ultrashort optical pulse, down to a resolution of 10 μm in space and 200 fs in time. Moreover, we directly show that the spatial phase information is not lost in the process, thanks to the coherence of the non-linear interaction. This implies that the concept of second-harmonic holography can be employed down to the sub-picosecond time scale, and used to discuss the features of the technique in terms of the reconstructed fields. © 2003 Elsevier B.V. All rights reserved.

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1. Introduction

The study of ultrafast phenomena has been a major scientific priority during the last decades covering different topics such as the study of radiation-matter interactions [1], transient response of molecules and atoms [2], coherent control of chemical reactions [3] or communication and information technology [4]. The growth of these fields relies upon the development of sources of femtosecond radiation and of appropriate techniques able to provide time domain information in the femtosecond scale. However, during the interaction of short optical pulses with a non-linear medium, different mechanisms can lead to its reshaping into complex spatio-temporal structures with non-trivial light distribution [5]. As a consequence, their complete characterization requires a
method capable of acquiring a snap-shot of their intensity distribution in the whole three-dimensional (3D; \(x, y, t - z/c\)) comoving frame.

Most of the available methods for pulse diagnostic provide information of the wave-packet (WP) characteristics in a space of reduced dimensionality. The use of frequency resolved autocorrelation techniques (FROG) or spectral shearing interferometry (SPIDER) allows for the recovery of the temporal intensity and phase profile of a given pulse but assumes uniform transverse spatial distribution [6,7]. On the contrary, the characterization of transversally localized beams often relies upon the optical imaging onto CCD cameras, therefore the temporal information is lost because of their integration times unavoidably larger than the optical pulse duration. Recently, a space–time characterization method based on improved SPIDER technique has been developed, capable of resolving electric field characteristics in time and along one spatial coordinate [8]. A quite direct way of obtaining spatio-temporal intensity profiles of a WP is to perform measurements with a streak camera, which allows a temporal resolution up to fractions of ps [9]. This technique allowed the investigation of the dynamics of the breakup along the pulse envelope of a large elliptical beam propagating into a saturable Kerr non-linear medium [10,11]. However, also in this case, the space–time maps are intrinsically two dimensional (1 spatial + 1 temporal dimensions).

A different approach to the problem considers the retrieval of the pulse shape through an all-optical processing by means of spatially resolved detection systems combined with gating techniques. The principle of the method is that of characterizing with spatial resolution an optical field that is proportional to the product \(E_O(x,t)E_R(x,t)\), where \(E_O(x,t)\) is the object to be measured and \(E_R(x,t)\) is a suitable reference pulse. Since the product is different from zero only on the intersection of the support of both fields, \(^2\) by translating the reference with respect to the object, we get the possibility of recording information from different parts of the object. Among the linear time gating techniques, light-in-flight holography has been the first technique which permitted the recording of dynamically evolving light fields during propagation [12–14]. Recently, this technique has been adapted to study the propagation of a 3 ps long pulse in linear media [15]. Linear probing techniques were also exploited in order to obtain time resolved imaging, like the probing of the birefringence properties of plasma by means of a delayed, spatially extended 100 fs pulses to investigate the dynamics of laser pulse focusing in air [16].

Nonlinear processes have been employed since long ago to resolve in time the evolution of ultrafast phenomena. Among them, the quadratic non-linearity has been proved to be particularly versatile due to the fact that it easily provides terms containing the product of two optical fields. Recently, a type II degenerate parametric amplification scheme has been employed to obtain time resolved 2D images of a picosecond-pulse hitting a diffusing screen with 35 ps resolution [17], thus yielding a 3D imaging. The same technique was later used to image an object embedded in a thick diffusing sample [17,18]. Although our setup is actually an improved version of that described in [18], we point out that our conceptual approach is different from the study of the propagation of a wave front. In fact, since we're recovering the intensity profile at the given position in space, in our case the propagation variable \(z\) is fixed.

Our goal in this article is to demonstrate the potentiality of the optical gating technique to acquire a high resolution space–time intensity map of short, focused WPs in their comoving reference frame. We propose a method that is based on quadratic type-I interaction in a sum-frequency generation scheme either by a non-collinear second harmonic generation or by a collinear sum-frequency (SF) scheme. The latter has been used in [19]. Here we discuss the first option, showing that if the angle between the two interacting fields is small enough, then a reliable space–time map of the object pulse can be obtained. This can be achieved if the duration of the gate is much smaller than that of the object to be imaged. A holo-

\(^2\) Rigorously, the support of a function is the set of points where its value is different from zero. For realistic optical fields that have exponentially decaying tails we can define a “practical” support, defined as the set of points in the space–time in which the field amplitude is larger than \(1/e\) times the peak value.
graphic interpretation of the method permits to gain insight into the process of up-conversion of the space–time slices of the object into the SF field, and to prove that the phase information is not lost in the imaging process, thanks to the coherence of the SF process. Our results confirm this possibility. The theoretical discussion of the method is followed by Section 3, where we present the set-up and the experimentally reconstructed space–time intensity profiles of a parametric spatial soliton excited by a 1 ps light pulse. For our setting, we estimate a mapping resolution of 200 fs in time and about 10 μm in space. The experimental confirmation of the holographic properties of the time-resolved images are also reported. The features of the technique are presented in Section 4, pointing out the limitations that may arise and discussing the possible implementations in each case. In the last section the main conclusions are presented.

2. Description of the technique: intensity and field reconstruction

In this section we explicitly show how a non-collinear sum-frequency scheme can be exploited to get high resolution space–time intensity maps of an unknown light wave packet with a space–time structure (object wave). The recovery of a 3D intensity map is obtained by means of a short reference pulse which provides a time gating inside a non-linear (NL) crystal, and generates a SF signal containing the information about a set of 2D slices of the object obtained by changing the reference delay. We first discuss the case for the reconstruction of the object intensity profile, and then we show how the intrinsic coherence of the SF process allows in fact for a truly holographic recording of the unknown object.

2.1. 3D Intensity profile mapping

Let us denote the object \( E_O \) and reference \( E_R \) wave packets as follows:

\[
E_O = E_O(x, y, z) e^{i \omega_0 t - k_z (\omega_0) z - k_x (\omega_0) x} + \text{c.c.},
\]

\[
E_R = E_R(x, y, z, t) e^{i \omega_2 t - k_x (\omega_2) x - k_z (\omega_2) z} + \text{c.c.},
\]

where the complex functions \( E_O(x, y, t, z) \) and \( E_R(x, y, t, z) \) are the slowly varying envelopes of two waves with frequencies \( \omega_1 \) and \( \omega_2 \). Note that the equations describe two wavepackets propagating in the positive \( z \)-direction and colliding at an angle \( \theta = 2 \arctan(k_z/k_x) \) in the \( x-z \) plane (here \( k = \sqrt{k_x^2 + k_z^2} = 2\pi/\lambda_0 \)). For a SF generation process occurring inside a quadratic non-linear crystal, the polarization source giving rise to the SF can be written as:

\[
P_{SF} \propto 2E_O E_R e^{i \omega_3 t - k_z (\omega_3) z} + \text{c.c.}
\]

where the phase and energy matching conditions \( k_z (\omega_1) = k_z (\omega_2) + k_x (\omega_2) \), \( k_x (\omega_1) = -k_x (\omega_2) \) and \( \omega_3 = \omega_1 + \omega_2 \) have been used. The SF field propagates along \( z \)-direction and has a slowly varying envelope function that we will indicate by \( E_{SF}(x, y, t, z) \). Now we introduce the following assumptions: (1) small depletion of both the \( E_O \) and \( E_R \) fields; (2) negligible diffraction and dispersion within the crystal; (3) equal group velocities of the object, reference and sum-frequency pulses, namely \( u_O, u_R \) and \( u_{SF} \), that is \( u_O = u_R = u_{SF} = u \). Note that all these assumptions approximately hold as long as the thickness of the crystal \( \Delta z \) is small compared to the characteristic lengths of the system (non-linear length, dispersion and diffraction lengths, pulse walkoff length). These assumptions allow to find a travelling reference frame for all the propagating pulses by introducing the retarded time \( \tau = t - z/u \). The general partial differential equations describing the interaction process then reduces to an ordinary differential equation for the envelope \( E_{SF} \). If we also introduce a time delay \( \tau_i \) on the reference wavefront, the equation takes the form:

\[
\frac{dE_{SF}(x, y, \tau, z)}{dz} = i2\sigma E_O(x, y, \tau, z) E_R(x, y, \tau - \tau_i, z),
\]

where \( \sigma \) is the non-linear coupling term. The last equation is readily integrated and, if the mixing crystal is placed at position \( z_0 \), it reads:

\[
E_{SF}(x, y, \tau, z_0) = i\sigma 2E_O(x, y, \tau, z_0) E_R(x, y, \tau - \tau_i, z_0) \Delta z.
\]
A deeper discussion about the meaning of this expression will be given in the following subsection. Here we just point out how the intensity profile of the SF field can be used to retrieve the 3D intensity map of the object. More precisely, for a given lag time \( \tau_i \), the spatially dependent SF fluence profile (the CCD signal) \( S(x,y,\tau_i, z_0) \) recorded just at the exit face of the mixing crystal is given by:

\[
S(x,y,\tau_i, z_0) \approx (\sigma \Delta z)^2 \int_{-\infty}^{+\infty} |E_0(x,y,\tau, z_0)|^2 \\
\times |E_R(x,y,\tau - \tau_i, z_0)|^2 d\tau.
\]

This expression provides the convolution between the intensity profiles of the object and reference wave packets, lagged in time by \( \tau_i \), and shows that the signal recorded by a CCD sensor is a linear function of the intensity of both the object and reference wavepackets. In the particular case in which the reference wave is spatially homogeneous in the transverse \( x-y \) plane, and temporally much shorter than the object, we can write expression (6) in the following form:

\[
S(x,y,\tau_i, z_0) \propto (\sigma \Delta z)^2 |E_R|^2 |I_0|^2,
\]

where \( I_0 = |E_0|^2 \).

By imposing a set of delays (\( \tau_i, i = 1, \ldots, n \)) to the reference WP with respect to the object, a reliable 3D reconstruction of the WP structure can be achieved by the collection of the \( n \) images \( S(x,y,\tau_i, z_0) \). By changing the plane \( z_0 \), the temporal evolution of the WP can also be obtained.

Notice that the model described above does not take into account the dispersion of the mixing crystal. Therefore, the model as it is does not predict limits to the resolution of the map as long as arbitrary short reference pulses are available. Actually the real mixing crystal has a finite bandwidth that limits the spatiotemporal resolution of the obtainable maps. Therefore a careful evaluation of the dispersion characteristics of the mixing crystal have to be gauged as ultrashort pulses are either investigated or used as a reference. More details are discussed in Section 4.

2.2. Holographic properties

As Eq. (5) clearly shows, the SF field is proportional to the field of the object WP. This point can also be explained in terms of a holographic description of the process, thus bringing to a deeper understanding of the imaging reconstruction process.

As stated in [20,23,24], for a plane wave reference the generated wavefront through the NL interaction behaves as a conventional hologram recorded at frequency \( \omega \) and illuminated with a radiation at \( 2\omega \). As a consequence, the position, scale, resolution and all the other properties of transformation of the reconstructed image can be predicted by means of the ordinary laws of holography. This hologram is recorded and reconstructed at a time, and exists only when light propagates inside the crystal. Yet we also point out that we obtain the 3D map of our pulse by collecting a set of independent 2D holograms by slicing the object pulse at different delays. Nevertheless, according to the holographic properties of the SF process, the slicing can be done with the mixing crystal at any \( z \) from the real object to be recovered and the information recorded is enough to reconstruct the slice.

In order to clarify this point, let’s assume to perform an experiment in which we reconstruct a slice of an object WP (Fig. 1(a)). At any distance \( z \) (Fig. 1(b)) the reconstructed WP will correspond to the propagated version of the one at \( z_0 \). Let us consider the case when the object WP has a bandwidth small enough with respect to the SF bandwidth so that our simplified model applies. Moreover we assume that the pulse does not have appreciable angular dispersion.\(^3\) Under these assumptions, any slice recorded in the confocal configuration (see Fig. 1(a)) could also be reconstructed when the mixing crystal is displaced at position \( z \) and the field to be converted is the propagated one. This is possible by exploiting the holographic properties, provided that the detecting system is set to reconstruct the virtual image of the slice. This image is placed at a distance \( 2z \) far from the mixing crystal (see Fig. 1(c)), its transverse size being identical to that of the object (see [23, and

\(^3\)In the case of angular dispersion, the diffraction drives an effective group velocity dispersion also in the vacuum, therefore the spatial evolution cannot be decoupled from the temporal one (see for example [21,22]).
also note that we are working in the degenerate case, $\omega_1 = \omega_2$.

The possibility to recover the intensity profile of a virtual image comes from the complete wavefront (field) reconstruction arising from the SF process coherence and contained in Eq. (4). 4

This important property also allows to get intensity maps with a wider dynamical range. For example, in the cases when the object is so intense that the undepleted pump approximation does not hold, we can get the WP profile by displacing the NL crystal to a plane where the intensity is reduced and then recover the intensity profile at the desired plane $z_0$ by suitably moving the imaging system (Fig. 1(c)).

3. Experimental results

We prepared several experiments in order to prove the possibility to recover 3D maps of short WPs and to show their holographic properties. The experimental set-up is sketched in Fig. 2.

The 1 ps pulses of a Nd:glass laser source (TWINKLE, Light Conversion, wavelength 1055 nm) are splitted on two lines by means of a beam splitter. In the first line, the laser pulse is frequency doubled in a KDP crystal and focused on a 15-mm long lithium triborate (LBO) crystal. For pulses about 1 \( \mu \)J in energy, spatial solitons are formed in this crystal because of the optical parametric generation process [25]. The gaussian temporal profile causes the spatial soliton to be formed only in the central part of the pulses where the intensity is higher [26]. This leads to a non-trivial space–time structure in the output pump wave packet [27], which we will consider as the object. Furthermore, the object has been magnified four times by means of a two-lens telescope imaging the LBO exit face into the NL mixing crystal (a 100 \( \mu \)m thick, $\beta$-barium borate crystal, BBO). The beam expansion has been necessary to: (i) reduce the beam intensity and therefore fulfill the undepleted mixing requirement; (ii) to avoid information loss related to the finite spatial resolution of the mixing process. In the second line, a 200-fs reference pulse is produced at the wavelength of 527.5 nm by means of a second-harmonic pulse compressor [28] and

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4 We suggest for example the possibility to use an interferometric or heterodyne device in order to obtain a complete characterization of each slice. By measuring the field instead of the intensity, a remarkable increment of the dynamical range is also possible.
expanded three times by means of a telescope. Both lines are recombined in the thin BBO crystal, cut and oriented to generate the non-collinear SF from the object and reference beams. An external incidence angle of ~6.5° between the propagation directions has been chosen. The delay between the two pulses can be varied by means of a suitable delay line placed on the reference pulse line. The non-collinear SF radiation is spatially selected by an aperture, then selected in frequency by means of coloured filters and the plane of the BBO is imaged onto the CCD sensor (PULNIX TM6CN). Finally, both the BBO crystal and the imaging system could move independently on a rail along the optical axis.

At first we focused the object inside the BBO crystal and adjusted the position of the imaging system as in Fig. 1(a). By recording the spatial profile of the SF radiation as a function of the reference pulse time lag (steps of $\Delta \tau_i = 66.6 \text{ fs}$ have been used, where $\Delta \tau_i = \tau_i - \tau_{i-1}$), we have retrieved the 3D isointensity maps of the object pulse at the exit face of the LBO crystal.

Fig. 3(a) shows three different intensity levels of the pulse in the $(x, y, \tau)$ space, while the corresponding contour plot of the $(x, \tau)$ plane section is depicted in Fig. 3(b). The space–time maps clearly show that the pulse is formed by a spatially focused structure followed by a diffracted tail. The accuracy of this reconstruction has been successfully tested by comparing the experimental plots with the results of a 3D-numerical simula-
tion of the formation process of the object pulse (see [27]).

In order to check for the validity of the holographic interpretation in this process, we have reconstructed the same WP of Fig. 3 by moving the NL crystal a distance $z$ far from the previous position and by scanning the position of the imaging system (CCD + lens) along the SF propagation direction in order to find out the position of the (virtual) reconstructed image. First we fixed the time lag between the object and the reference and selected a slice of the object corresponding to a narrow focused spot of $\sim 40$ $\mu$m in diameter inside the BBO crystal (corresponding to case a in Fig. 1). Furthermore, the displacements $z$ of the BBO crystal have been chosen large enough to ensure that the propagated wavefronts had lost the transverse structure. As a rough estimate, for the focused part of the object, the Rayleigh range is about 7 mm long, while we spanned distances from $-15$ to 15 mm. In Fig. 4, data show the position of the virtual image against the position of the real object from the crystal (see Fig. 1(c)), showing a remarkable fit to a straight line which slope is close to the value of 2, according to what discussed in the previous section. The error bars shown in the figure indicate the estimated uncertainties in the reconstructed image plane position, measured by scanning the whole Rayleigh range and by extracting the position of the central point. In the same figure the two insets show two isointensity maps recovered for the case when the imaging system is focusing directly the BBO plane ($z = 0$) and when the BBO crystal has been moved a distance $z = 15$ mm (the maximum propagation distance we imposed to the BBO). The agreement between the two intensity profiles proves the reliability of our method to work with virtual images in recovering 2D slices of WPs like those we used here.

In order to verify that we have always been operating in the undepleted regime, we have measured the dependence of the generated SF field as a function of the object-pulse energy at $z = 0$. The results are presented in Fig. 5, where the peak fluence (as registered from the CCD images) is plotted vs the transmission of the neutral-density filters that attenuate the object. The constant slope confirms the absence of any saturation in the SF process. Data also indicate that a slight overestimate of the background has been done during the profile acquisition (zero SF field is found for 15% filter transmission).

4. Features of the technique

In the previous sections we have proved that the described technique is able to perform the 3D
intensity mapping of objects similar to those we used for our measurements. Anyway, one can expect that in general the reliability of the technique to recover the slices could be affected by particular effects dealing with the interaction geometry, the mixing crystal, and the features of the interacting pulses, in particular for broad-band, ultra-short and chirped pulses. Therefore we discuss here three main features of our technique, namely, the duplication bandwidth of the mixing crystal, the interaction angle and the reference pulse shape, that could limit the fidelity of the technique.

4.1. Space–time resolution

As we briefly mentioned above, the interaction model we describe does not take into account the actual finite bandwidth of the mixing crystal, limiting the spatio-temporal resolution of the maps. In fact, in our experiment the BBO crystal has been chosen thin enough that the converted bandwidth was large compared to the object one. As a matter of fact, the spatio-temporal resolution of the slices is dictated by the maximum range of angles and frequencies over which the conversion is effective (for very broad-band objects the angular blurring that could arise from the phase matching condition should be considered). The key parameter here is the maximum phase mismatch between the fundamental and the SF waves at which the conversion efficiency vanishes, $\Delta k_{\text{max}}$. On the basis of the existing literature [29], we have estimated the maximum bandwidth converted by the crystal as the FWHM of the efficiency curve, yielding to a temporal bandwidth of about $630 \text{ cm}^{-1}$ and an angular bandwidth of about $510 \text{ cm}^{-1}$. By considering only the angles for which the conversion efficiency is higher than $1/e$ of the maximum value [30], we obtain the remarkable resolution of details approximately $10 \mu m$ in size (100 lines/mm). We can notice that the holographic interpretation of the wavefront reconstruction leads to an easy description of the imaging process. Furthermore, our optical system could be used in principle to achieve a microscopy of objects much shorter than the one we used here [19], although the limit of very short WP is detrimental for the holographic reconstruction of the phases.

4.2. Interaction angle

A point to be discussed concerns the influence of the non-collinear geometry and pulse chirp on the reconstruction of the hologram. Actually, the holographic interpretation is strictly valid for collinear geometry only (when $k_2 = 2k_1$) [23]. Although this condition is not strictly fulfilled in our experiment, we have maintained the interaction angle small enough to make this disturbance negligible. However, a temporal chirp in the object pulse could give rise to a spatial phase distortion of the SF wavefront [31]. Although this does not affect the results obtained with the confocal configuration (see Fig. 1(a)), it could distort the maps obtained in the holographic one (Fig. 1(c)). We expect that this effect is really rampant only when complicated ultrashort pulses with strong chirp are considered, or large interaction angle are employed. We are confident that our system was operated far from this limit, since, as we checked from the data, our experimental holographic maps do not show any appreciable enhancement of the astigmatism.

As it is well known, the non-collinear SF scheme is largely used to get single-shot autocorrelation traces of ultrashort pulses [32]. In fact, because of the geometry of the interaction, it is easy to show that the time of the interaction between the object and reference pulses depends on the transverse coordinate of the intersecting planes (see [2, p. 426]). Therefore, if the interacting angle is large, we expect that distortions of the spatial profile along this direction could affect the space–time maps. However, it turns out that in the confocal configuration (i.e., as in Fig. 1(a)) the effect of the interaction angle is merely that of skewing the intensity map, so that the actual time-axis of the object pulse is not parallel to that measured on the delay line. This is evident in the map depicted in Fig. 6, obtained with an external crossing angle of $\sim16^{\circ}$.

In the paraxial approximation, the slope of the object–pulse time-axis on the space–time map is given by:
\[ \tan \beta = \frac{c}{n} \sin \frac{\gamma}{2}, \]  

(8)

where \( \gamma \) is the angle between the propagation directions of the object and the reference wavepackets inside the crystal, and \( n \) is the refractive index of the medium. It is easy to recognize this formula as the calibration relation of the non-collinear, single-shot intensity autocorrelator [2,32].

### 4.3. Reference pulse shape

Finally, the last point to be discussed concerns the shape of the reference pulse. In our experiments it was only slightly affected by the presence of satellites because of the generation via the pulse compressor [28], the amplitude of these structures being small enough to be considered negligible to our aim. Actually, the recovered intensity profile of the object would be distorted in case the reference pulse has a structure more complicated than the single peak envelope, as Eq. (6) clearly points out. This suggests that a careful measurement of the reference shape (for example carried out by means of an autocorrelation technique) can be used to retrieve the real intensity profile by means of a deconvolution procedure. We point out, however, that any deconvolution unavoidably introduces an extra spurious noise which could degrade the final quality of the mapping [33].

## 5. Conclusions

We have shown that the 3D intensity maps of optical WPs with a complex space–time structure can be retrieved by an optical gating technique. The method allows the reconstruction of the WP in its comoving reference frame. The maps are obtained by suitably imaging the second harmonic radiation obtained by cross-correlating an object pulse with a much shorter plane wave-packet delayed in time, by means of a slightly non-collinear, degenerate SF process. The holographic properties of the generated radiation have been carefully tested experimentally, and the distortions of the intensity maps introduced by the non-collinear geometry have been discussed in detail. Finally, theoretical considerations point out that the ultimate resolution of our set-up is in the order of 100 lines/mm in space, and of a few femtoseconds in time. However, the choice of the reference pulse limits the actual time resolution to about 200 fs.

We foresee that the developed technique will benefit all the fields where a space–time mapping of light pulses is relevant, such as the investigations on the reshaping of ultrashort pulses propagating in non-linear materials [5,10,11,19,27]. Moreover, the holographic features of the technique might be exploited to fully reconstruct the field of an object pulse and its evolution during the propagation.

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