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Duration of ultrashort pulses in the presence of spatio-temporal coupling

C. Bourassin-Bouchet,^{1,*} M. Stephens,^{1,2} S. de Rossi,¹ F. Delmotte,¹
and P. Chavel¹

¹Laboratoire Charles Fabry, Institut d'Optique, CNRS, Univ Paris-Sud, 2 avenue Augustin
Fresnel, 91127 Palaiseau cedex, France

²Physics Department, University of Michigan, Ann Arbor, Michigan 48109, USA

[*charles.bourassin-bouchet@institutoptique.fr](mailto:charles.bourassin-bouchet@institutoptique.fr)

Abstract: We report on a simple method allowing one to decompose the duration of arbitrary ultrashort light pulses, potentially distorted by space-time coupling, into four elementary durations. Such a decomposition shows that, in linear optics, a spatio-temporal pulse can be stretched with respect to its Fourier limit by only three independent phenomena: nonlinear frequency dependence of the spectral phase over the whole spatial extent of the pulse, spectral amplitude inhomogeneities in space, and spectral phase inhomogeneities in space. We illustrate such a decomposition using numerical simulations of complex spatio-temporal femtosecond and attosecond pulses. Finally we show that the contribution of two of these three effects to the pulse duration is measurable without any spectral phase characterization.

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References and links

1. E. D. Potter, J. L. Herek, S. Pedersen, Q. Liu, and A. H. Zewail, "Femtosecond laser control of a chemical reaction," *Nature* **355**, 66–68 (1992).
2. W. Boutu, S. Haessler, H. Merdji, P. Breger, G. Waters, M. Stankiewicz, L. J. Frasinski, R. Taïeb, J. Caillat, A. Maquet, P. Monchicourt, B. Carré, and P. Salières, "Coherent control of attosecond emission from aligned molecules," *Nat. Phys.* **4**, 545–549 (2008).
3. I. Walmsley, L. Waxer, and C. Dorrer, "The role of dispersion in ultrafast optics," *Rev. Sci. Instrum.* **72**, 1–29 (2001).
4. C. Fiorini, C. Sauteret, C. Rouyer, N. Blanchot, S. Seznec, and A. Migus, "Temporal aberrations due to misalignments of a stretcher-compressor system and compensation," *IEEE J. Quantum Electron.* **30**, 1662–1670 (1994).
5. K. Osvay, A. P. Kovács, Z. Heiner, G. Kurdi, J. Klebniczki, and M. Csatari, "Angular dispersion and temporal change of femtosecond pulses from misaligned pulse compressors," *IEEE J. Quantum Electron.* **10**, 213–220 (2004).
6. X. Gu, S. Akturk, and R. Trebino, "Spatial chirp in ultrafast optics," *Opt. Commun.* **242**, 599–604 (2004).
7. Z. Bor, "Distortion of femtosecond laser pulses in lenses," *Opt. Lett.* **14**, 119–121 (1989).
8. U. Fuchs, U. D. Zeitner, and A. Tünnermann, "Ultra-short pulse propagation in complex optical systems," *Opt. Express* **13**, 3852–3861 (2005).
9. M. Kempe, U. Stamm, B. Wilhelmi, and W. Rudolph, "Spatial and temporal transformation of femtosecond laser pulses by lenses and lens systems," *J. Opt. Soc. Am. B* **9**, 1158–1165 (1992).
10. T. A. Planchon, S. Ferré, G. Hamoniaux, G. Chériaux, and J.-P. Chambaret, "Experimental evidence of 25-fs laser pulse distortion in singlet beam expanders," *Opt. Lett.* **29**, 2300–2302 (2004).
11. C. Bourassin-Bouchet, S. de Rossi, F. Delmotte, and P. Chavel, "Spatiotemporal distortions of attosecond pulses," *J. Opt. Soc. Am. A* **27**, 1395–1403 (2010).

12. S. Akturk, X. Gu, P. Gabolde, and R. Trebino, "The general theory of first-order spatio-temporal distortions of Gaussian pulses and beams," *Opt. Express* **13**, 8642–8661 (2005).
13. S. Akturk, X. Gu, P. Bowlan, and R. Trebino, "Spatio-temporal couplings in ultrashort laser pulses," *J. Opt.* **12**, 093001 (2010).
14. P. Gabolde, D. Lee, S. Akturk, and R. Trebino, "Describing first-order spatio-temporal distortions in ultrashort pulses using normalized parameters," *Opt. Express* **15**, 242–252 (2007).
15. S. Akturk, X. Gu, E. Zeek, and R. Trebino, "Pulse-front tilt caused by spatial and temporal chirp," *Opt. Express* **12**, 4399–4410 (2004).
16. Y. Mairesse, A. de Bohan, L. J. Frasinski, H. Merdji, L. C. Dinu, P. Monchicourt, P. Breger, M. Kovacev, R. Taïeb, B. Carré, H. G. Muller, P. Agostini, and P. Salières, "Attosecond synchronization of high-harmonic soft x-rays," *Science* **302**, 1540–1543 (2003).
17. Z. L. Horváth, A. P. Kovács, and Z. Bor, "Distortion of ultrashort pulses caused by aberrations," in *International Conference on Ultrafast Phenomena (UP)*, 2006 OSA Technical Digest Series (Optical Society of America, 2006), paper ThD16.
18. C. Dorrer, E. M. Kosik, and I. A. Walmsley, "Direct space-time characterization of the electric fields of ultrashort optical pulses," *Opt. Lett.* **27**, 548–550 (2002).
19. P. Gabolde and R. Trebino, "Single-shot measurement of the full spatio-temporal field of ultrashort pulses with multispectral digital holography," *Opt. Express* **14**, 11460–11467 (2006).
20. P. Bowlan, U. Fuchs, R. Trebino and U. D. Zeitner, "Measuring the spatiotemporal electric field of tightly focused ultrashort pulses with sub-micron spatial resolution," *Opt. Express* **16**, 13663–13675 (2008).
21. B. Alonso, I. J. Sola, O. Varela, J. Hernández-Toro, C. Méndez, J. San Román, A. Zaïr, and L. Roso, "Spatiotemporal amplitude-and-phase reconstruction by Fourier-transform of interference spectra of high-complex-beams," *J. Opt. Soc. Am. B* **27**, 933–940 (2010).
22. L. Gallmann, G. Steinmeyer, D. H. Sutter, T. Rupp, C. Iaconis, I. A. Walmsley, and U. Keller, "Spatially resolved amplitude and phase characterization of femtosecond optical pulses," *Opt. Lett.* **26**, 96–98 (2001).
23. I. Walmsley and C. Dorrer, "Characterization of ultrashort electromagnetic pulses," *Adv. Opt. Photon.* **1**, 308–437 (2009).
24. P. Mercère, P. Zeitoun, M. Idir, S. Le Pape, D. Douillet, X. Levecq, G. Dovillaire, S. Bucourt, K. A. Goldberg, P. P. Naulleau, and S. Rekawa, "Hartmann wave-front measurement at 13.4 nm with $\lambda_{EUV}/120$ accuracy," *Opt. Lett.* **28**, 1534–1536 (2003).
25. C. Dorrer, and I. A. Walmsley, "Simple linear technique for the measurement of space-time coupling in ultrashort optical pulses," *Opt. Lett.* **27**, 1947–1949 (2002).

1. Introduction

The generation, manipulation and characterization of ultrashort light pulses have become an area of considerable interest in the past few decades. Such pulses allow probing ultrafast phenomena such as chemical reactions [1] using femtosecond pulses, or the electronic motion in molecules [2] thanks to the decrease of the pulse duration down to the attosecond time scale. One major assumption in ultrafast optics is that time and space are uncoupled variables, which means that an ultrashort pulse can be described as the product of a time dependent pulse shape with a space dependent beam profile [3]. It considerably simplifies the way to describe a pulse since the pulse shape is supposed to be same in every point in space.

However, since the beginning of ultrafast science, it has been shown that this assumption is often invalid. For example, misaligned prisms or gratings compressors, or aberrated optical systems can spatio-temporally distort a pulse [4–10], which causes a coupling between time and space variables. Moreover, as shorter and shorter pulses become accessible to experiment, this coupling has become more and more difficult to avoid. For example, it has been shown that attosecond extreme ultraviolet (XUV) pulses are very sensitive to optical aberrations of typical focusing mirrors [11]. So there is an increasing need for theoretical tools to describe the coupling phenomenon.

Especially, associating a duration to a pulse is not straightforward anymore. Indeed, since coupling makes the pulse change in space, its duration has to be locally defined [12, 13]. Nevertheless, it remains important to be able to summarize the full spatio-temporal pulse into a single duration. To do so, it has been proposed to consider the duration of the spatially integrated pulse, namely the global pulse duration [9, 10, 12, 13]. Some theoretical models studying

the global duration already exist [12–14] for both gaussian temporal and spatial profiles and for first order coupling.

In this article, we propose a more general analysis of the global duration of arbitrary pulses. More precisely, we report on a way to decompose the root mean square (RMS) global pulse duration of a spatio-temporal pulse into four basic durations based on the behavior of the complex spectrum versus frequency and space. Each of these durations has a simple physical explanation. Firstly, we describe the principle of the duration decomposition. As an illustration, we single out each of them in a simple situation that may arise in the femtosecond regime, and we proceed with a example in the attosecond regime. Finally, we show that the contributions of three of the four elementary durations to the global pulse duration are measurable without any spectral phase characterization.

2. Theoretical Study

A linearly polarized light pulse propagating in vacuum can be considered as a scalar electric field E solution of the propagation equation, that is Eq. (1), where z stands for the position along the pulse propagation axis, x and y are the transverse coordinates and t is the time variable:

$$\nabla^2 E(x, y, z, t) - \frac{1}{c^2} \frac{\partial^2 E(x, y, z, t)}{\partial t^2} = 0 \quad (1)$$

If considering the problem at a given position z_0 , one typically assumes that the spatio-temporal electric field (resp. the spatio-spectral electric field) can be written [3] as a product of a space-dependent function $g(x, y)$ with a time-dependent function $f(t)$ (resp. a frequency-dependent function $\tilde{f}(\omega)$), where \tilde{f} stands for the Fourier Transform of f), see Eq. (2):

$$E(x, y, z_0, t) = g(x, y) \cdot f(t) \quad (2)$$

Defining a duration for such a pulse is straightforward since it suffices to estimate the characteristic width of the intensity function $|f|^2$. One possible way to get such a width is to calculate the RMS duration Δt defined by Eq. (3):

$$\Delta t^2 = \langle t^2 \rangle - \langle t \rangle^2 \quad (3)$$

where $\langle \rangle$ stands for the usual mean operator weighted by $|f|^2$, so that $\langle t \rangle$ is equal to $\int_{-\infty}^{+\infty} |f(t)|^2 t dt / \int_{-\infty}^{+\infty} |f(t)|^2 dt$.

When developing Eq. (3), we get Eq. (4) which is well-established [3]:

$$\Delta t^2 = \Delta t_{FT}^2 + \Delta GD^2 \quad (4)$$

In this equation, GD is the *Group Delay* and is equal to $d\varphi/d\omega$, with φ the spectral phase of the pulse. According to Eq. (4), the duration Δt depends on two parameters:

- (i) Δt_{FT} is the *Fourier Transform limited* duration. It corresponds to the duration of a pulse, the GD of which is constant with respect to frequency. It is the shortest RMS duration attainable with a given spectrum.
- (ii) ΔGD is the RMS spectral variation of the GD. It quantifies the temporal synchronization of the spectral components.

To summarize, any spectral variations of the GD will stretch an ultrashort pulse, according to Eq. (4).

However, these results are based on the major assumption that the pulse is not distorted by any space-time coupling, see Eq. (2), which is a strong restriction. In order to establish a

more general formula for the duration of an arbitrary spatio-temporal pulse, we first have to find a way to convert a space-time signal into a temporal one, whose duration will be easier to estimate. As proposed in [9, 10], the simplest way to do so is to spatially integrate the pulse. The resulting temporal signal, hereafter referred to as the *Global Pulse* $I_G(t)$, would be the signal detected by an imaginary photodiode with a suitable temporal resolution but no spatial resolution. Moreover, the detection plane of such a sensor would be orthogonal to the pulse propagation axis, see Eq. (5):

$$I_G(t) = \iint_{-\infty}^{+\infty} |E(x, y, t)|^2 dx dy \quad (5)$$

By analogy, one can define global quantities in the spectral domain, such as the *Global Spectrum* as the spatially integrated spectrum $S_G(\omega)$, and the *Global Group Delay* as the spatially averaged group delay $GD_G(\omega)$, see Eqs. (6) and (7):

$$S_G(\omega) = \iint_{-\infty}^{+\infty} |\tilde{E}(x, y, \omega)|^2 dx dy \quad (6)$$

$$GD_G(\omega) = \langle GD \rangle_{(x,y)} = \iint_{-\infty}^{+\infty} |\tilde{E}(x, y, \omega)|^2 GD dx dy / \iint_{-\infty}^{+\infty} |\tilde{E}(x, y, \omega)|^2 dx dy \quad (7)$$

where $\langle \rangle_{(x,y)}$ is the mean weighted by the spatial intensity. Hereafter, it will be convenient to also use the spectral weighted mean $\langle \rangle_{(\omega)}$. Moreover, the combination of the spatial mean with the spectral mean will be summarized into the operator $\langle \rangle_{(x,y,\omega)}$.

Applying Eq. (3) to the temporal signal $I_G(t)$, we get the *global pulse duration* Δt_G , as named in [12, 13], that is the RMS duration of the spatially integrated pulse. This duration can be written the following way:

$$\Delta t_G^2 = \Delta t_{FTG}^2 + \Delta GD_G^2 + \tau_{AC}^2 + \tau_{PC}^2 \quad (8)$$

The complete demonstration of such a decomposition is given in Appendix A. According to Eq. (8), the duration Δt_G involves four parts:

- (i) Δt_{FTG} is the *Global Fourier Transform limited duration*. It is the shortest RMS global duration attainable with the involved spectral components. It corresponds to the duration of a temporal pulse $I_{FTG}(t)$, the spectrum of which is the global spectrum $S_G(\omega)$, and the spectral phase of which is zero. This fundamental limit is reached if there is no spectro-spatial coupling and the spectral phase is linear, see Fig. 1(a).
- (ii) ΔGD_G is the *Dispersion of the Global Group Delay*. It represents the global synchronization of the spectral components for a spatio-temporal pulse, as shown on Fig. 1(b). This term stands for phenomena such as temporal chirp or higher order dispersion. When there is no space-time coupling, the GD does not depend on (x, y) anymore, so ΔGD_G reduces to ΔGD , which was previously defined in Eq. (4).
- (iii) τ_{AC} represents the temporal stretch due to a coupling of the spatio-spectral amplitude. Hereafter, such a coupling will be referred to as an *Amplitude Coupling* and τ_{AC} as the *Amplitude Coupling Duration*, see Fig. 1(c). This duration is zero if every spectral component is equally distributed spatially, that is if the local Fourier-Transform limited duration $\Delta t_{FT}(x, y)$ is equal to Δt_{FTG} in every point in space, see Eq. (9). τ_{AC} includes the influences of phenomena such as spatial chirp.

$$\tau_{AC} = \langle \Delta t_{FT}^2(x, y) - \Delta t_{FTG}^2 \rangle_{(x,y)}^{1/2} \quad (9)$$

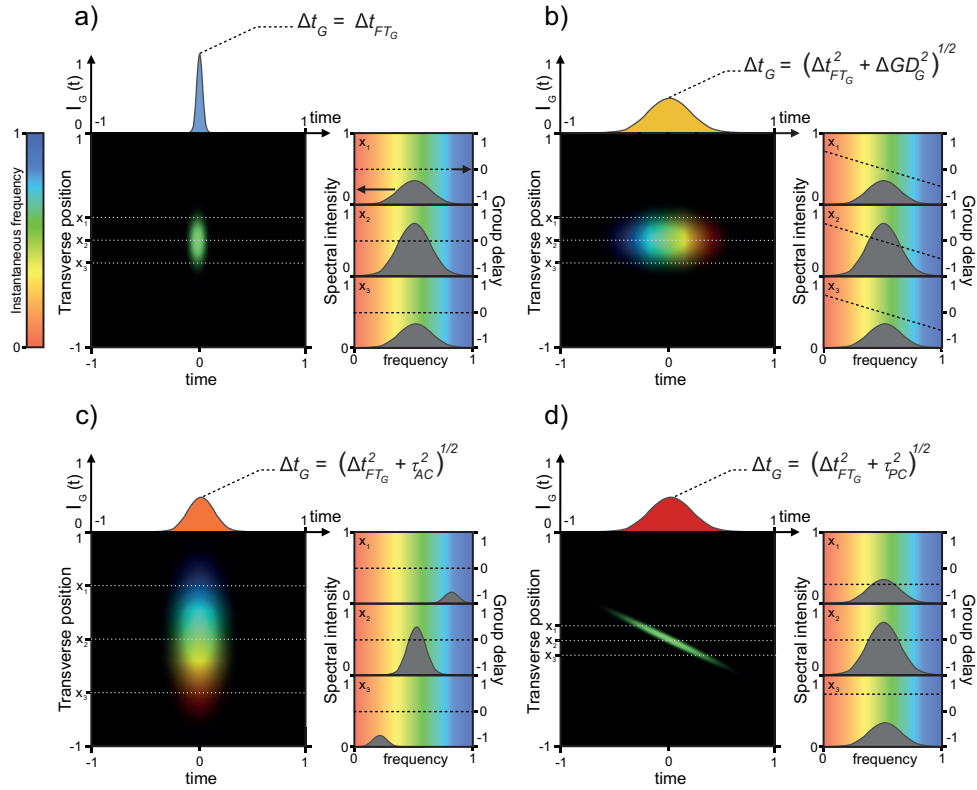


Fig. 1. Description of the decomposition of the global duration of an arbitrary pulse into four elementary durations. The scales are in arbitrary units. For each case, the instantaneous frequency of the spatio-temporal pulse (central part) and its corresponding global pulse (on top) are depicted, along with the local spectrum (gray shaded lines) and group delay (black dashed line) for three positions (on the right part). (a) The shortest pulse duration attainable Δt_{FTG} is reached when every spectral component is synchronized both in space and time and is equally spread in space. The global pulse will be stretched by (b) any temporal desynchronization of the spectral components, due to a temporal chirp, for example, (c) any inhomogeneity in their spatial spread, induced by phenomena such as spatial chirp, or (d) any spatial desynchronization of these spectral components, caused by spatially varying time delay, for example.

- (iv) τ_{PC} stands for the *Phase Coupling Duration*, that is the temporal stretch induced by a coupling on the spatio-spectral phase. It quantifies the spatial synchronization of every spectral component, see Eq. (10) and Fig. 1(d). This has an influence when there is a spatio-spectral coupling in the phase, *i.e.* when the phase cannot be written as a *sum* of a frequency-dependent function with a space-dependent function. It summarizes the effects of phenomena such as spatially varying time delay.

$$\tau_{PC} = \left\langle [GD(x, y, \omega) - GD_G(\omega)]^2 \right\rangle_{(x, y, \omega)}^{1/2} \quad (10)$$

Equation (4) highlighted that a pulse without space-time coupling can be described as a Fourier-transform limited pulse stretched by any non linear variations of its spectral phase. Equation (8) extends this analysis to spatio-temporal pulses. It shows that any space-time coupling can be seen as a *deviation of local properties from global properties* (see Eqs. (9), and (10)), and that any coupling stretches a pulse. Moreover, Eq. (8) is a general formula which is accurate for *arbitrarily* complex spatio-temporal pulses.

3. Examples of Decomposition of the Duration

3.1. Basic Examples

In order to see the influence of these four parameters on distorted pulses, we now consider several examples of duration decomposition. We numerically simulate a simple pulse with a zero spectral phase and no space-time coupling, namely a 800nm gaussian pulse with a 10fs RMS duration. The pulse spatial profile is Gaussian and its RMS width Δx is equal to $1\mu\text{m}$. Then we add various distortions on the pulse, and numerically calculate the duration decomposition in each case. To simplify the understanding of the described pulses, we here consider only (x, t) dependent pulses, so the previous equations remain valuable if removing the y coordinate.

First, let us consider a pulse that is distorted by a time delay which linearly varies in space with a slope γ equal to $20\text{fs}/\mu\text{m}$. We obtain the pulse reported on Fig. 2(a), the pulse front of which is tilted. The numerically obtained duration decomposition highlights that the pulse stretch is due to phase coupling. Indeed, there is no amplitude coupling since the spectrum remains the same in every point in space. This distortion only affects the group delay, the latter being given by Eq. (11):

$$GD(x) = \gamma \cdot (x - x_0) \quad (11)$$

In this equation, x_0 is the central radial position of the pulse, which is equal to $0\mu\text{m}$. Since the group delay does not depend on ω , all the spectral components remains synchronized at a given point in space, so that GD_G is zero. On the other hand, a spatially varying GD indicates a phase coupling. Indeed, using Eq. (10) gives immediately Eq. (12):

$$\tau_{PC} = |\gamma| \cdot \Delta x \quad (12)$$

Given that Δx is equal to $1\mu\text{m}$, Eq. (12) predicts a pulse stretch equal to 20fs , as shown on Fig. 2(a).

Now, consider the situation where the pulse is distorted by both spatial and temporal chirps. The spatial chirp is characterized by a spatial shift ζ of the spectral components of $-0.118\mu\text{m}/\text{nm}$, and the *Group Delay Dispersion* (GDD) inducing the temporal chirp is equal to $400\text{fs}^2/\text{rad}$. It is well established that adding temporal and spatial chirp creates pulse front tilt [15], which is visible on Fig. 2(b). According to our duration decomposition, it appears that

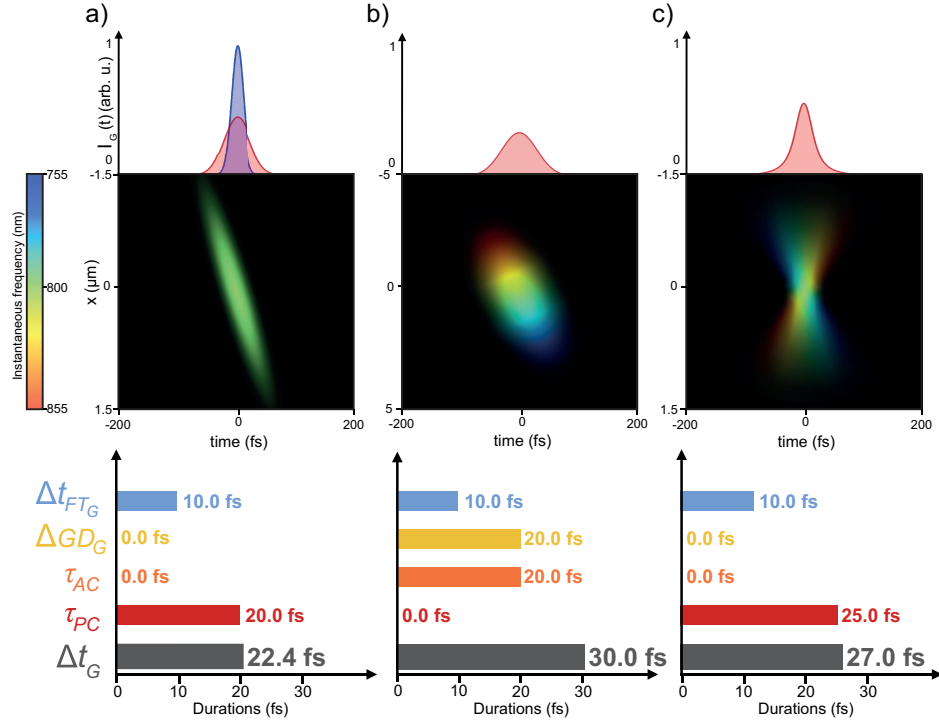


Fig. 2. Three examples of the duration decomposition of spatio-temporal pulses. The spatio-temporal pulses and the global pulses (red shaded curves) are reported on the upper panels. The associated duration decompositions are reported on the lower panels. (a) Pulse front tilt caused by a time delay linearly varying in space. (b) Pulse front tilt caused by both spatial and temporal chirps. (c) Pulse distorted by a space dependent Group Delay Dispersion (negative for negative positions, and positive for positive ones). The Global Fourier Transform limit $I_{FT_G}(t)$ of the three distorted pulses is reported in a) (blue shaded curve).

the global pulse is equally stretched by ΔGD_G and amplitude coupling. Indeed, the GDD modifies the spectral phase while leaving the spectrum unchanged, as shown by Eq. (13), so it can only modify ΔGD_G or τ_{PC} :

$$GD(\omega) = GDD \cdot (\omega - \omega_0) \quad (13)$$

where ω_0 equals 800nm and represents the central angular frequency of the spectrum. Since the group delay is constant in space, $GD(x, y, \omega)$ equals $GD_G(\omega)$ in every point, so that τ_{PC} is zero whereas ΔGD_G is given by Eq. (14):

$$\Delta GD_G = |GDD| \cdot \Delta\omega \quad (14)$$

This expression stands for the usual stretch term of an uncoupled pulse distorted by a constant GDD [3]. As for spatial chirp, it spatially spreads the spectral components, so that the spectrum becomes narrower than $S_G(\omega)$ at a given point in space, as depicted on Fig. 1(c). Consequently, τ_{AC} cannot be null insofar as the local Fourier Transform limited duration $\Delta t_{FT}(x)$ becomes greater than Δt_{FT_G} . Indeed, it was shown [6] that adding a linear spatial chirp to a pulse, the beam and pulse profile of which are Gaussian, makes the local width of the spectrum $\Delta\omega'(x)$ decrease, see Eq. (15):

$$\Delta\omega' = \left(\frac{1}{\Delta\omega^2} + \frac{\zeta^2}{\Delta x^2} \right)^{-1/2} \quad (15)$$

where $\Delta\omega$ is the spectrum width without spatial chirp. Moreover, it is well-known that the quantity $\Delta t_{FT} \cdot \Delta\omega'$ is equal to $1/2$ for gaussian pulses. This allows to deduce Δt_{FTG} and the local Fourier Transform limited duration $\Delta t_{FT}(x)$, see Eqs. (16) and (17):

$$\Delta t_{FTG}^2 = \frac{1}{4\Delta\omega^2} \quad (16)$$

$$\Delta t_{FT}^2 = \langle \Delta t_{FT}^2 \rangle_{(x)} = \frac{1}{4\Delta\omega'^2} = \frac{1}{4\Delta\omega^2} + \frac{\zeta^2}{4\Delta x^2} \quad (17)$$

Thus, using the definition of the amplitude coupling duration, we obtain τ_{AC} :

$$\tau_{AC} = \frac{|\zeta|}{2\Delta x} \quad (18)$$

According to Eq. (14) and (18), both ΔGD_G and τ_{AC} have to be equal to $20 fs$. Finally, by comparing Figs. 2(a) and 2(b), it appears that pulse front tilt, or more generally distortion of the shape of the pulse front, can be obtained either by adjusting the phase coupling, or by choosing a combination of amplitude coupling with a nonzero global group delay.

As a third example, let us consider a radially varying GDD characterized by a parameter ξ , which equals $500 fs^2/rad/\mu m$. To be more specific, the pulse depicted on Fig. 2(c) is distorted by a positive (resp. negative) GDD for positive (resp. negative) positions and zero on the propagation axis. The resulting global pulse is clearly stretched by these distortions. But according to the duration decomposition, the global pulse is not chirped since ΔGD_G remains null. Indeed, since the GDD is alternatively negative and positive in different points in space, $GD_G(\omega)$ is zero. In other words, the spectral components remain temporally synchronized on average. Finally, the real source of pulse stretching is phase coupling, insofar as the GD depends on space. More precisely, the group delay is given by Eq. (19), and the corresponding phase coupling duration by Eq. (20):

$$GD(x, \omega) = \xi \cdot (\omega - \omega_0) \cdot (x - x_0) \quad (19)$$

$$\tau_{PC} = |\xi| \cdot \Delta x \cdot \Delta\omega \quad (20)$$

The latter gives a value of τ_{PC} equal to $25 fs$, as shown on Fig. 2(c).

3.2. Complex Example

To go one step further with this analysis of the duration decomposition, we now consider more complex spatio-temporal pulses. Optical aberrations are known to be a typical source of spatio-temporal distortions of ultrashort pulses [7–9]. In particular, it has been shown that attosecond XUV pulses are very sensitive to aberrations induced by most focusing mirrors [11]. Moreover, such pulses are chirped due to the generation process [16]. The latter phenomenon, called the atto-chirp, is unavoidable. So the attosecond pulses can be stretched in a highly complex way due to a combination of atto-chirp and optical aberrations. Now we will investigate the influence of these various phenomena by decomposing the global duration of such distorted attosecond pulses.

To do so, we simulate an attosecond pulse reflected off a grazing incidence ellipsoidal mirror, and we compare the aberration-free case with a configuration that is aberrated due to misalignment. The focal length of the mirror is equal to $750 mm$ and its optimal grazing angle is equal

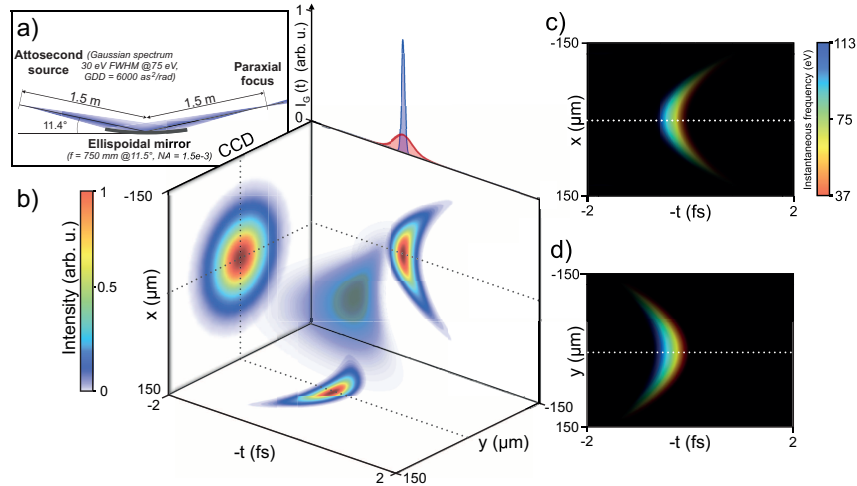


Fig. 3. Simulation of the full spatio-temporal electric field distribution of an attosecond pulse distorted by optical aberrations. (a) The attosecond pulse is focused by a grazing incidence ellipsoidal mirror, the optimal grazing angle of which is equal to 11.5° . The mirror is set with a grazing angle of 11.4° to see the impact of astigmatism on the pulse. (b) The spatio-temporal intensity distribution of the pulse at the paraxial focus is represented. The projections on the (x, t) and (y, t) sides of the box stand for two slices of the 3D pulse located on the dashed lines. The projection on the (x, y) side represents the temporally integrated pulse, that is the image that should be obtained if using a CCD sensor. The pulses $I_G(t)$ (red shaded curve) and $I_{FG}(t)$ (blue shaded curves) associated to the spatio-temporal pulse are represented above the box. (c) (resp. (d)) The instantaneous frequency of the (x, t) (resp. (y, t)) projection is plotted. Astigmatism is responsible for the inverted curvatures of the pulse front on the two projections, whereas the atto-chirp makes the instantaneous frequency vary linearly throughout the pulse envelope.

to 11.5° , leading to sagittal and tangential radii of curvature equal to 1500mm and 299mm , respectively. To add some aberrations to the pulse, the mirror is set in the focus-focus configuration, but with a grazing angle reduced to 11.4° , see Fig. 3(a).

The attosecond pulse generated at the source is chosen not to be distorted by space-time coupling. Its XUV spectrum has a gaussian envelope, centered at 75eV with a full width at half maximum (FWHM) of 30eV . The intrinsic GDD has a typical value of $6000\text{as}^2/\text{rad}$ [16], and the divergence of the beam is between 1 and 2mrad over the whole spectrum. Since we simulate a grazing incidence mirror, we can consider that its reflectivity is constant and its spectral phase is linear over the whole spectrum, which is the case for gold or platinum made mirrors. The theoretical model for the simulations is described in [11]. As shown in Fig. 3(b), the full (x, y, t) pulse is simulated. This becomes necessary to completely see the influence of aberrations that clearly depend both on x and y . To be more specific, Fig. 3(b) reports on the evolution in space and time of the intensity of the attosecond pulse at the paraxial focus of the ellipsoidal mirror. The obtained spatio-temporal pulse is clearly distorted by astigmatism [17], leading to a stretch of the global pulse. In addition to astigmatism, the atto-chirp increases the pulse duration too. As shown on Figs. 3(c) and 3(d), this causes the instantaneous frequency to vary throughout the envelope of the pulse.

To separate the influences of the atto-chirp and of the aberrations, we study the evolution of the pulse and of its duration decomposition during the transition from an aberration free case,

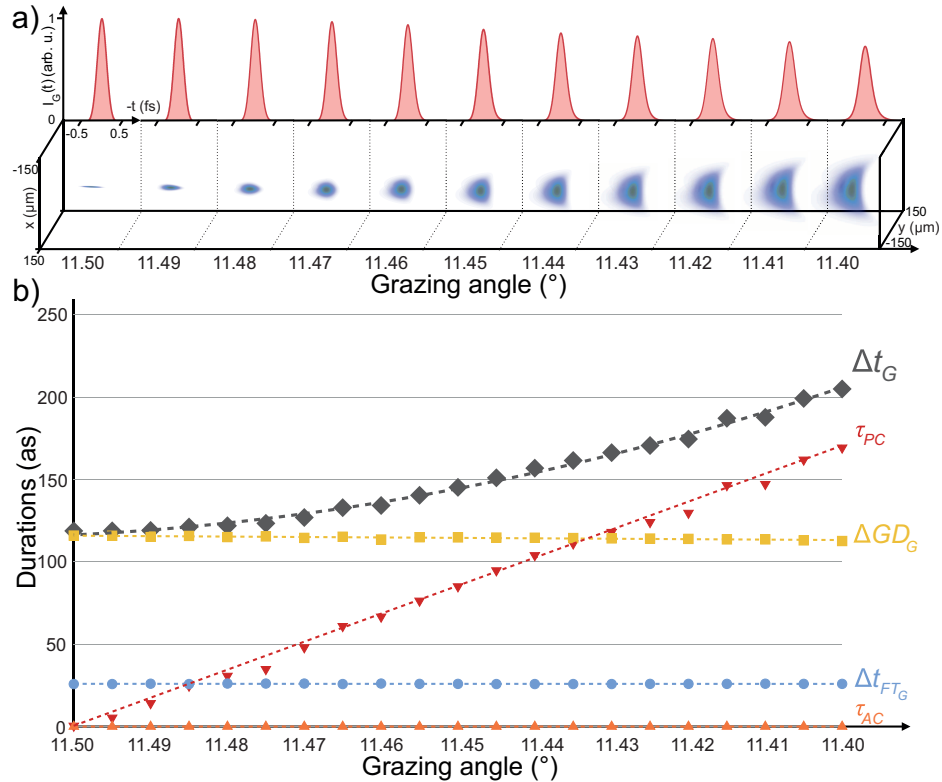


Fig. 4. Evolution of the duration decomposition of a refocused attosecond pulse with respect to the strength of astigmatism, that is with respect to the deviation from the optimal grazing angle of 11.5° to 11.4° . (a) Simulation of the spatio-temporal pulses (lower part) and their global pulses (upper part) with respect to the grazing angle. (b) Evolution of the global duration of the pulse (black diamonds) and its decomposition into the four parameters: Δt_{FTG} (blue circles), ΔGD_G (yellow squares), τ_{AC} (orange up triangles), τ_{PC} (red down triangles), and their corresponding fits (dashed lines).

i. e. where the only possible stretch phenomenon is the atto-chirp, to the previous aberrated case. We first consider that the mirror is set at its optimal grazing angle of 11.5° , leading to a diffraction limited pulse, see Fig. 4(a). At this angle, the duration of the obtained global pulse is equal to 118 as (278 as FWHM). The decomposition shows that Δt_{FTG} is equal to 26 as , see Fig. 4(c). This duration is also the Fourier Transform limited pulse duration given by Eq. (16) using the initial gaussian spectrum. Indeed the global spectrum does not change after the reflection off the mirror since its reflectivity was supposed to be constant over the whole spectrum. Moreover, the spectral phase of the grazing incidence mirror was assumed to be linear due to the total reflection phenomenon. So the obtained ΔGD_G of 115 as also corresponds to the temporal stretch of a pulse, the spectrum and GDD of which are the initial gaussian spectrum and the initial GDD, as confirmed by Eq. (14). Finally it appears that the two coupling durations are zero, which is consistent since a diffraction limited pulse at its focus is not distorted by any space-time coupling.

We now consider the evolution of the decomposition while changing the grazing angle from 11.5° down to 11.4° , *i. e.* while adding astigmatism to the attosecond pulse. Δt_{FTG} remains

constant since geometric aberrations do not change the involved spectral components, that is $S_G(\omega)$. As for ΔGD_G , it appears that the global group delay $GD_G(\omega)$ does not vary either, since the atto-chirp remains constant whatever the strength of aberrations. Moreover, the amplitude coupling duration remains null whereas τ_{PC} increases linearly with respect to the grazing angle. Indeed, as it was in the case of Fig. 3(c) and 3(d), the pulse is composed of the same spectral components in every point in space, so the spectrum does not vary in space. But the major consequence of astigmatism is to curve the pulse front by applying a time delay depending on space to the diffraction limited pulse. This phenomenon was already described on Fig. 1(d) and Fig. 2(a), and is known to be pure phase coupling. So the stronger the astigmatism, the greater the phase coupling duration. Moreover, it appears that a 0.1° misalignment of the mirror is sufficient to almost double the global pulse duration, which confirms the high sensitivity of attosecond pulses to optical aberrations.

These numerical simulations confirm that the duration decomposition is suitable to describe various, realistic and potentially highly complex spatio-temporal pulses.

4. Experimental Considerations and Discussion

For now, the duration decomposition has only been used to theoretically describe spatio-temporal pulses. But it could be interesting to know how to measure these durations experimentally. Of course, if we use techniques able to characterize the full spatio-temporal electric field of a pulse [18–21], the four durations can be easily extracted. But some other methods, which are easier to implement, may in fact give access to other measures of duration. Moreover, since the elementary durations correspond to four independent phenomena, these techniques are not necessarily the same for all the durations:

- i) Δt_{FTG} is given by the spatially integrated spectrum $S_G(\omega)$. So a typical spectrometer without spatial resolution is sufficient to get $I_{FTG}(t)$ and its duration.
- ii) τ_{AC} can be obtained by measuring the spatially resolved spectrum with an imaging spectrometer [6].
- iii) ΔGD_G can be determined applying interferometric spectral phase characterization techniques such as Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER). Indeed, when using such techniques, the retrieved group delay is usually known up to a constant [22, 23] which can depend on position in presence of phase coupling. Consequently, performing independent measurements in different points of the pulse does not allow to reconstruct the full spatio-temporal pulse. Nevertheless, it does not prevent from determining ΔGD_G , since this space dependent constant GD does not play any role in the global group delay. More precisely, the experimental group delay GD_{xp} can be considered as a centered group delay $GD(x_0, y_0, \omega) - \langle GD \rangle_{(\omega)}(x_0, y_0)$ where the absolute GD at the point (x_0, y_0) has been lost. According to Eq. (21), ΔGD_G depends only on GD_{xp} . So knowing the latter is sufficient to determine ΔGD_G . Nevertheless, it should be noted that the mean of the GD has to be weighted by the spatially-resolved spectrum, meaning that the spatio-spectral intensity has to be extracted from the measurements.

$$\Delta GD_G = \left\langle \left(\langle GD \rangle_{(x,y)} - \langle GD \rangle_{(x,y,\omega)} \right)^2 \right\rangle_{(\omega)}^{1/2} = \left\langle \langle GD_{xp} \rangle_{(x,y)}^2 \right\rangle_{(\omega)}^{1/2} \quad (21)$$

- iv) τ_{PC} can be measured using a simple wavefront sensor. More precisely, a Shack-Hartmann-like wavefront sensor can measure the shape of monochromatic wavefronts

but not the phase relation between these wavefronts, that is the spectral phase. However, to do a similar analysis as in the case of ΔGD_G , the measured wavefront $\varphi_{xp}(x, y, \omega_0)$ at the ω_0 frequency can be seen as a centered wavefront $\varphi(x, y, \omega_0) - \langle \varphi \rangle_{(x,y)}(\omega_0)$ without any spectral phase information. Thus it becomes simple to get the phase coupling duration by performing spectrally resolved wavefront measurements, see Eq. (22).

$$\tau_{PC} = \left\langle \left(GD - \langle GD \rangle_{(x,y)} \right)^2 \right\rangle_{(x,y,\omega)}^{1/2} = \left\langle \left(\frac{\partial \varphi_{xp}}{\partial \omega} \right)^2 \right\rangle_{(x,y,\omega)}^{1/2} \quad (22)$$

Therefore, it appears that the four durations can be measured using independent experimental techniques and various devices, such as an imaging spectrometer, a SPIDER-like system or a wavefront sensor. In particular, the latter is usually able to measure both the intensity and the wavefront of a beam. So if the sensor is coupled with a tunable filter for the spectral resolution, it would allow to also access the spatially resolved spectrum, *i. e.* to measure Δt_{FG} , τ_{AC} and τ_{PC} at the same time. Moreover, Hartmann wavefront sensors exist for the XUV range [24], so such a setup would be potentially usable for XUV attosecond pulses if coupled with selective XUV mirrors to ensure the spectral resolution. Another way to access the space-time coupling without spectral phase measurements was given in [25]. The proposed solution is based on two spatially sheared and phase-shifted replicas of a pulse sent into an imaging spectrometer. By doing so, one can measure the spatially resolved spectrum along with the variations of the spectral phase across the beam, and thus get Δt_{FG} , τ_{AC} and τ_{PC} . These simple setups could be a good way to measure and minimize both amplitude and phase coupling without the need for complete spatio-temporal characterization techniques.

5. Conclusion

We described a way to summarize the complexity of *arbitrary* spatio-temporal light pulses into four elementary durations, namely one fundamental limit and three extra stretch terms. These three terms highlight the influence of three independent stretch phenomena on spatio-temporal pulses, named the dispersion of the global group delay, the amplitude coupling and the phase coupling. We illustrated this decomposition using numerical simulations of femtosecond and attosecond pulses distorted by various phenomena, such as pulse front tilt or optical aberrations. Moreover, we discussed a possible way to experimentally measure these durations, and considered simple setups to retrieve three of these terms without spectral phase characterization. We conclude that this duration decomposition appears to have the potential to become a useful tool for describing and characterizing ultrashort light pulses.

6. Appendix A: Duration of an Arbitrary Spatio-Temporal Pulse

We consider the RMS duration Δt_G of the spatially integrated pulse, namely the *Global pulse* $I_G(t)$:

$$\Delta t_G^2 = \langle t^2 \rangle - \langle t \rangle^2 = K^{-1} \int_{-\infty}^{+\infty} I_G(t) t^2 dt - \left(K^{-1} \int_{-\infty}^{+\infty} I_G(t) t dt \right)^2 \quad (23)$$

where K stands for a normalization constant and is equal to $\int_{-\infty}^{+\infty} I_G(t) dt$.

6.1. Calculation of $\langle t \rangle^2$

In order to develop Eq. (23), we first calculate $\langle t \rangle^2$, which leads to Eq. (24):

$$\begin{aligned}
\langle t \rangle &= \frac{\int_{-\infty}^{+\infty} I_G(t) t dt}{\int_{-\infty}^{+\infty} I_G(t) dt} = K^{-1} \iiint_{-\infty}^{+\infty} |E(x, y, t)|^2 t dt dx dy \\
&= K^{-1} \iiint_{-\infty}^{+\infty} E^*(x, y, t) \cdot E(x, y, t) t \cdot dt dx dy
\end{aligned} \tag{24}$$

where $E^*(x, y, t)$ corresponds to the complex conjugate of $E(x, y, t)$. Using Parseval's theorem, which gives Eq. (25):

$$\langle t \rangle = -iK^{-1} \iiint_{-\infty}^{+\infty} \tilde{E}^*(x, y, \omega) \frac{\partial \tilde{E}(x, y, \omega)}{\partial \omega} dx dy d\omega \tag{25}$$

where $\tilde{E}(x, y, \omega)$ represents the Fourier transform of $E(x, y, t)$ and is equal to $|\tilde{E}(x, y, \omega)| \exp(i\varphi(x, y, \omega))$. Hereafter $|\tilde{E}(x, y, \omega)|$ (resp. $\varphi(x, y, \omega)$) will be written $|\tilde{E}|$ (resp. φ). Moreover, since $\langle t \rangle$ is a real quantity, we obtain Eq. (26):

$$\begin{aligned}
\langle t \rangle &= -iK^{-1} \iiint_{-\infty}^{+\infty} |\tilde{E}| \exp(-i\varphi) \left[\frac{\partial |\tilde{E}|}{\partial \omega} + i |\tilde{E}| \frac{\partial \varphi}{\partial \omega} \right] \exp(i\varphi) dx dy d\omega \\
&= 0 + K^{-1} \iiint_{-\infty}^{+\infty} |\tilde{E}|^2 \frac{\partial \varphi}{\partial \omega} dx dy d\omega
\end{aligned} \tag{26}$$

If noticing that K is equal to $\iiint_{-\infty}^{+\infty} |\tilde{E}|^2 d\omega dx dy$, we get Eq. (27):

$$\langle t \rangle = \frac{\iint_{-\infty}^{+\infty} \left(\int_{-\infty}^{+\infty} |\tilde{E}|^2 d\omega \right) \langle GD \rangle_{(\omega)} dx dy}{\iint_{-\infty}^{+\infty} \left(\int_{-\infty}^{+\infty} |\tilde{E}|^2 d\omega \right) dx dy} = \left\langle \langle GD \rangle_{(\omega)} \right\rangle_{(x, y)} = \langle GD \rangle_{(x, y, \omega)} \tag{27}$$

Moreover, we can decompose $\langle t \rangle^2$, leading to Eq. (28):

$$\begin{aligned}
\langle t \rangle^2 &= \langle GD \rangle_{(x, y, \omega)}^2 = \left\langle \langle GD \rangle_{(x, y)} \right\rangle_{(\omega)}^2 + \left\langle \langle GD \rangle_{(x, y)}^2 \right\rangle_{(\omega)} - \left\langle \langle GD \rangle_{(x, y)}^2 \right\rangle_{(\omega)} \\
&= \left\langle \langle GD \rangle_{(x, y)}^2 \right\rangle_{(\omega)} - \Delta GD_G^2
\end{aligned} \tag{28}$$

6.2. Calculation of $\langle t^2 \rangle$

We still have to calculate $\langle t^2 \rangle$ in order to get Δt_G . Using Parseval's theorem leads to Eq. (29):

$$\begin{aligned}
\langle t^2 \rangle &= K^{-1} \int_{-\infty}^{+\infty} I_G(t) t^2 dt = K^{-1} \iiint_{-\infty}^{+\infty} \left[\left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 + |\tilde{E}|^2 \left(\frac{\partial \varphi}{\partial \omega} \right)^2 \right] dx dy d\omega \\
&= K^{-1} \iiint_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 dx dy d\omega + \langle GD^2 \rangle_{(x, y, \omega)}
\end{aligned} \tag{29}$$

Using the definition of the global spectrum $S_G(\omega)$, see Eq. (6), one can establish Eq. (30):

$$\left(\frac{\partial (S_G(\omega))^{1/2}}{\partial \omega} \right)^2 = \frac{1}{4S_G(\omega)} \left(\frac{\partial S_G(\omega)}{\partial \omega} \right)^2 = \frac{1}{4S_G(\omega)} \left(\iint_{-\infty}^{+\infty} \frac{\partial |\tilde{E}|^2}{\partial \omega} dx dy \right)^2$$

$$= \frac{1}{S_G(\omega)} \left(\iint_{-\infty}^{+\infty} |\tilde{E}| \frac{\partial |\tilde{E}|}{\partial \omega} dx dy \right)^2 \quad (30)$$

Moreover, using the Cauchy-Schwarz inequality allows to get inequality [Eq. (31)]:

$$\begin{aligned} \left(\iint_{-\infty}^{+\infty} |\tilde{E}| \frac{\partial |\tilde{E}|}{\partial \omega} dx dy \right)^2 &\leq \iint_{-\infty}^{+\infty} |\tilde{E}|^2 dx dy \cdot \iint_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 dx dy \\ &\Leftrightarrow \left(\frac{\partial (S_G(\omega))^{1/2}}{\partial \omega} \right)^2 \leq \iint_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 dx dy \end{aligned} \quad (31)$$

We can introduce the *Amplitude Coupling Duration* τ_{AC} which is a positive or null quantity homogenous to a duration:

$$K^{-1} \int_{-\infty}^{+\infty} \left(\frac{\partial S_G(\omega)^{1/2}}{\partial \omega} \right)^2 d\omega + \tau_{AC}^2 = K^{-1} \iiint_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 dx dy d\omega \quad (32)$$

It is easily checkable that τ_{AC} is null if $|\tilde{E}|$ can be written as a product of a spectral function with a space-dependent function, that is if there is no coupling on the spatio-spectral amplitude. Moreover:

$$K^{-1} \int_{-\infty}^{+\infty} \left(\frac{\partial S_G(\omega)^{1/2}}{\partial \omega} \right)^2 d\omega = K^{-1} \int_{-\infty}^{+\infty} I_{FTG}(t) t^2 dt = \Delta t_{FTG}^2 \quad (33)$$

where $I_{FTG}(t)$ is the *Global Fourier Transform limited pulse* which is, by definition, obtained by calculating the Fourier transform of the amplitude of $S_G(\omega)$ for a zero spectral phase. If noticing that the squared Fourier transform limited pulse duration at a given point $\Delta t_{FT}^2(x, y)$ is equal to $\int_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 d\omega / \int_{-\infty}^{+\infty} |\tilde{E}|^2 d\omega$, we get Eq. (34):

$$K^{-1} \iiint_{-\infty}^{+\infty} \left(\frac{\partial |\tilde{E}|}{\partial \omega} \right)^2 dx dy d\omega = \frac{\iint_{-\infty}^{+\infty} \left(\int_{-\infty}^{+\infty} |E|^2 dt \right) \Delta t_{FT}^2(x, y) dx dy}{\iint_{-\infty}^{+\infty} \left(\int_{-\infty}^{+\infty} |E|^2 dt \right) dx dy} = \langle \Delta t_{FT}^2(x, y) \rangle_{(x, y)} \quad (34)$$

Using Eqs. (29), (32), (33) and (34) leads to Eq. (35):

$$\langle t^2 \rangle = \Delta t_{FTG}^2 + \tau_{AC}^2 + \langle GD^2 \rangle_{(x, y, \omega)} \quad (35)$$

where τ_{AC} is equal to $\langle \Delta t_{FT}^2(x, y) - \Delta t_{FTG}^2 \rangle_{(x, y)}^{1/2}$.

Finally, using Eqs. (23), (28) and (35), one can get the RMS global pulse duration, see Eq. (36):

$$\Delta t_G^2 = \Delta t_{FTG}^2 + \tau_{AC}^2 + \langle \langle GD^2 \rangle_{(x, y)} \rangle_{(\omega)} - \langle \langle GD \rangle_{(x, y)}^2 \rangle_{(\omega)} + \Delta GD_G^2$$

$$= \Delta t_{FTG}^2 + \Delta GD_G^2 + \tau_{AC}^2 + \tau_{PC}^2 \quad (36)$$

where τ_{PC} is equal to $\left\langle \left\langle GD^2 \right\rangle_{(x,y)} - \left\langle GD \right\rangle_{(x,y)}^2 \right\rangle_{(\omega)}^{1/2}$, which can be rewritten $\left\langle \left(GD - GD_G \right)^2 \right\rangle_{(x,y,\omega)}^{1/2}$. It should be noticed that if there is no coupling on the spatio-spectral phase, that is if GD does not depend on space, τ_{PC} is null whatever the spectral phase.

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