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Tilted-Pulse Second-Harmonic Beam Analysis for Femtosecond to Subnanosecond Laser Pulse-Duration Measurements

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Abstract. The possibility of extending the second-harmonic beam (SHB) method proposed originally for picosecond and subpicosecond pulse-duration measurements to the femtosecond region is pointed out. This can be achieved by introducing a differential time delay of the pulse wave front corresponding to a tilting of the pulse in the direction other than that applied by Wyatt and Marinero, and also by Saltiel et al., who achieved extensions towards the subnanosecond region. The solution of the wave equations for noncollinear second-harmonic generation in the case of arbitrarily tilted pulses has been carried out. Simple formulae valid from the subnanosecond to the femtosecond region are presented.

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The second-harmonic beam (SHB) method proposed earlier for the investigation of temporal characteristics of short laser pulses [1, 2] was found to be very efficient for picosecond and subpicosecond pulses [3–5], and appears to be the easiest method for duration measurements of single pulses. The related autocorrelation method first proposed by Maier et al. [6] requires strict reproducibility of the pulses and extremely narrow beams for pulses shorter than 1 ps [1]. The very expensive streak camera seems to have attained its limit of applicability at ~ 1 ps. For reviews of methods for pulse duration measurements see [7, 8].

In the SHB method two replicas of the pulse to be measured are used to generate a SH beam in a noncollinear arrangement in a nonlinear crystal where the SH radiation is only produced in the region where the two replicas overlap both in space and time. For fundamental pulses having lengths (durations) of the order or larger than the fundamental beam width (which is limited by the crystal size) the SH beam width saturates and carries information on the fundamental beam width rather than on the pulse duration (Fig. 1a).

Recently, the SHB method has been extended beyond this saturation limit for the measurement of subnanosecond pulses by introducing a linear differential time delay along the fundamental wave front using either a grating [9, 10] or a Michelson echelon [11].

A differential time delay, i.e. a pulse delayed to a linearly varying extent at different points of its beam cross-section can be also described by a tilting of the front of the intensity envelope function and can be visualized by slanting (sliding askew) a pack of playing cards. As shown by a comparison of Fig. 1a and b a proper tilting of the "long" pulse, in fact, gives an extension towards longer pulses: there is a narrower SH beam width in the tilted case containing information on the pulse duration.

For short enough pulses one has no saturation problem, the width of the overlap region where SH is generated depends linearly on the duration of the

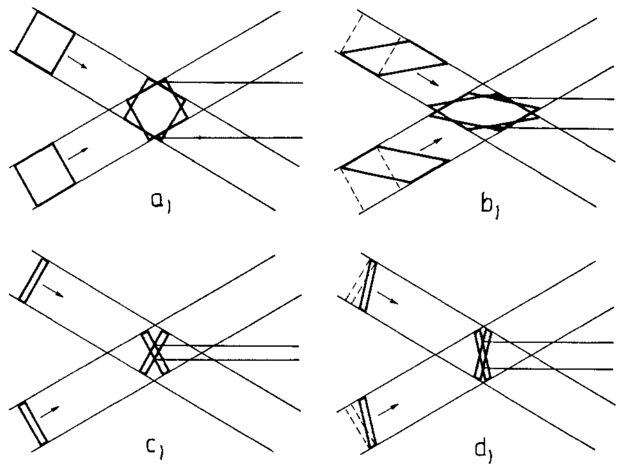

Fig. 1a–d. Limits of the SHB method for long (a) and short (c) pulses and extensions of the method by tilting the pulse in various directions [(b) and (d)]. The overlap regions are supposed to be inside of a nonlinear crystal and for the sake of simplicity rectangularly shaped pulses are shown

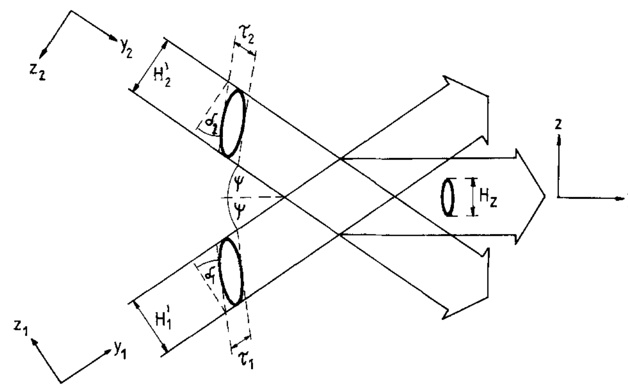

Fig. 2. Schematic arrangement of noncollinear SH generation of tilted Gaussian pulses inside a nonlinear crystal having its main axis parallel to the z-axis

fundamental pulse, moreover, one has a symmetrized time-to-space mapping of the pulse structure. For durations $\tau \lesssim 0.1$ ps, however, the overlap region becomes extremely narrow (Fig. 1c) and the SH beam width will be determined by diffraction and other side effects [2, 3, 12].

In the present paper we show how a differential time delay corresponding to a tilting of the pulse in the opposit direction, as applied by Wyatt and Marinero [9] and also by Saltiel et al. [11], can be used to extend the SHB method down to the femtosecond region. As seen on Fig. 1d such a tilt, in fact, results in a broader SH beam thus diminishing the role of side effects. The solution of the wave equations for tilted pulses is, in general, given resulting in simple formulae valid from the subnanosecond to the femtosecond region.

The General Case

We use the usual ooe arrangement for noncollinear SH generation (Fig. 2) applying, however, differentially delayed, i.e. tilted pulses. In the calculations we assume Gaussian pulses, accurate phase-matching and neglect absorption phenomena. For generality we start with differently processed replica pulses which may have different beam diameters, tilting angles and even durations. (E.g., in the case of refraction on a grating a beam of circular cross-section will be transformed into one of elliptical cross-section, in addition to tilting of the wave front.)

In this case the nonzero component of the electric field vectors inside the nonlinear crystal of the tilted fundamental pulses having a carrier frequency ω and wavenumber k can be written (for the role of a delay between pulses see [1])

$$E_{jx} = g_j(\mathbf{r}_j, t)\cos(\omega t - k y_j), \tag{1}$$

where $j = 1, 2$ and we use the cartesian coordinate systems x, y_j, z_j introduced separately for beams 1 and 2, y_j pointing into the propagation direction, z_j towards the other beam before the crossing. The envelope functions g_j have the form

$$g_j(\mathbf{r}_j, t) = E_{j0}\exp(-2\ln 2\{x^2/H_j^2 + z_j^2/H_j'^2 + [t - (y_j - z_j\tan\delta_j)/u]^2/\tau_j^2\}). \tag{2}$$

Here H_j and H_j' are beam width parameters (fwhm) in the x and z_j-directions, respectively, δ_j are the tilting angles, u is the group velocity of the ordinary fundamental pulses, and τ_j are the pulse durations (fwhm) without tilting (Fig. 2). The terms x^2/H_j^2 and $z_j^2/H_j'^2$ define the beams from which the term $[t - (y_j - z_j\tan\delta_j)/u]^2/\tau_j^2$ taylors the desired differentially delayed shape of the pulses. In the limit $\delta_j = 0$ we obtain the case without differential time delay described in [1]. It should be pointed out that the angles δ_j characterize the tilts of the pulses *inside* the crystal. Pulses tilted towards the x-direction will not be considered here resulting in no apparent advantages for measurements.

The extraordinary SH wave propagates perpendicularly to the main axis z of the crystal in the direction defined as the y-axis (Fig. 2). Assuming nondepletion of the fundamental waves the nonzero z component of the electric field vector of the SH wave can be found as the solution of the equation

$$\frac{\partial^2 E_z}{\partial y^2} = \frac{k^2\cos^2\psi}{\omega^2}\frac{\partial^2}{\partial t^2}(E_z + 4\pi d_{zxx}E_{1x}E_{2x}), \tag{3}$$

where d_{zxx} is the relevant SHG coefficient, 2ω and $2k\cos\psi$ are the frequency and the wave-vector of the SH wave, ψ is half of the crossing angle of the fundamental beams. We are looking for a solution of (3) in the form

$$E_z(\mathbf{r}, t) = f(\mathbf{r}, t)\sin[2(\omega t - ky\cos\psi)], \tag{4}$$

where f is the envelope function of the SH pulse. In the approximation of slowly varying amplitudes from (3) and (4) we have

$$\left(\frac{\partial}{\partial y} + \frac{k\cos\psi}{\omega}\frac{\partial}{\partial t}\right) f(\mathbf{r}, t) = -4\pi d_{zxx} k\cos\psi g_1 g_2 . \tag{5}$$

Introducing the new variable $\vartheta = t - ky\cos\psi/\omega$ instead of t and using throughout x, y, z and ϑ as independent variables we obtain the ordinary linear differential equation

$$\frac{d}{dy} f(\mathbf{r}, t(y, \vartheta)) = F\exp\left[-2\ln\left(2\sum_{u,v=x,y,z,\vartheta}\sigma_{uv}uv\right)\right], \tag{6}$$

where the constant F, together with the coefficients σ_{uv} in the bilinear exponent can be found by comparison of (6) with (5) and by using (2). The nonvanishing coefficients are

$$\sigma_{xx} = H_1^{-2} + H_2^{-2}, \tag{6a}$$

$$\sigma_{yy} = (H_1'^{-2} + H_2'^{-2})\sin^2\psi + p_1^2u^{-2}\tau_1^{-2} + p_2^2u^{-2}\tau_2^{-2}, \tag{6b}$$

$$\sigma_{zz} = (H_1'^{-2} + H_2'^{-2})\cos^2\psi + q_1^2u^{-2}\tau_1^{-2} + q_2^2u^{-2}\tau_2^{-2}, \tag{6c}$$

$$\sigma_{\vartheta\vartheta} = \tau_1^{-2} + \tau_2^{-2}, \tag{6d}$$

$$\sigma_{yz} = \sigma_{zy} = (-H_1'^{-2} + H_2'^{-2})\cos\psi\sin\psi - p_1q_1u^{-2}\tau_1^{-2} + p_2q_2u^{-2}\tau_2^{-2}, \tag{6e}$$

$$\sigma_{y\vartheta} = \sigma_{\vartheta y} = p_1u^{-1}\tau_1^{-2} + p_2u^{-1}\tau_2^{-2}, \tag{6f}$$

$$\sigma_{z\vartheta} = \sigma_{\vartheta z} = -q_1u^{-1}\tau_1^{-2} + q_2u^{-1}\tau_2^{-2}, \tag{6g}$$

where the notations

$$p_j = (nu/c - 1)\cos\psi + \tan\delta_j\sin\psi, \tag{6h}$$

$$q_j = \sin\psi - \tan\delta_j\cos\psi, \tag{6i}$$

have been used, n standing for the index of refraction of the ordinary fundamental wave. Equation (6) can be solved to obtain the envelope function f as a function of x, y, z, ϑ. The integration can be carried out trivially in the case if we are interested only in the SH wave leaving the crystal and the crystal is big enough to contain the whole of the intersection region of the pulses. In this reasonable approximation the integration limits can be extended to $y \to \pm\infty$, respectively, to have

$$f(x, z, \vartheta) \simeq G\exp\left\{-2\ln 2\left[\sum_{u,v=x,z,\vartheta}\sigma_{uv}uv - (\sigma_{yz}z + \sigma_{y\vartheta}\vartheta)^2/\sigma_{yy}\right]\right\}, \tag{7}$$

where G is a constant. This result can be used to calculate the distribution of the SH energy incident on the detector system defined as

$$W(x, z) = \frac{1}{4\pi}\int_{-\infty}^{\infty} E_z^2(\mathbf{r}, t)dt \simeq \frac{1}{8\pi}\int_{-\infty}^{\infty} f^2(x, z, \vartheta)d\vartheta . \tag{8}$$

Using (7) and (6a) we obtain the Gaussian distribution

$$W(x, z) = K\exp[-4\ln 2(x^2/H_x^2 + z^2/H_z^2)], \tag{9}$$

where K is a constant and

$$H_x = \sigma_{xx}^{-1/2} = (H_1^{-2} + H_2^{-2})^{-1/2}, \tag{9a}$$

$$H_z = [\sigma_{zz} + (\sigma_{yy}\sigma_{z\vartheta}^2 + \sigma_{yz}^2\sigma_{\vartheta\vartheta} - 2\sigma_{yz}\sigma_{y\vartheta}\sigma_{z\vartheta})/(\sigma_{y\vartheta}^2 - \sigma_{yy}\sigma_{\vartheta\vartheta})]^{-1/2}, \tag{9b}$$

are the easily measurable widths (fwhm) of the SH energy distribution, the latter containing according to (6b–6g) information about the pulse durations τ_j. The analysis of the result (9b) confirms the qualitative deductions one obtains from drawings similar to Fig. 1. The analysis will be carried out here in the symmetric case of identical replica since this is the one which can be used most effectively for extending the time domain of measurements.

The Symmetrical Case

The results (9a, b) can be considerably simplified in the case of a symmetric arrangement when $\delta_1 = \delta_2 = \delta$, $\tau_1 = \tau_2$, $H_1 = H_2$ and $H_1' = H_2'$. In fact, for this case we have $p_1 = p_2$, $q_1 = q_2$, $\sigma_{yz} = \sigma_{z\vartheta} = 0$ and $H_z = \sigma_{zz}^{-1/2}$. Accordingly the widths of the SH energy distribution (fwhm) along the x and z-axis defined in (9) can be written using (9a, b), (6c and i) in the form

$$H_x = H/\sqrt{2}, \tag{10a}$$

$$H_z = H_{z\infty}(1 + \tau_0^2/\tau^2)^{-1/2}, \tag{10b}$$

where τ is the duration and H the beam diameter in the x-direction of the fundamental pulses (fwhm),

$$H_{z\infty} = H'/(\sqrt{2}\cos\psi) \tag{11}$$

is the saturation value of H_z for long pulses, H' is the beam diameter (fwhm) of the fundamental pulses in the yz plane inside the crystal, ψ is shown on Fig. 2, and

$$\tau_0 = \tau_0(\delta) = H'|\tan\psi - \tan\delta|/u \tag{12}$$

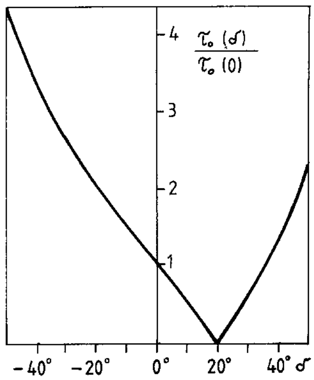

Fig. 3. The variation of the timescale constant τ_0 in (10b) and (12) as a function of the tilting angle δ in the symmetric case assuming $\psi = 19.7°$ which is the phase-matching angle in $LiIO_3$ for $\lambda = 1.06\,\mu m$

is a time scale constant characterizing the range of pulse durations available for measurement by the present method in a given experimental arrangement. The $\tau_0(\delta)$ function is plotted in Fig. 3. The relative errors of τ and H_z have the ratio

$$\frac{\Delta\tau}{\tau} \Big/ \frac{\Delta H_z}{H_z} = 1 + \tau^2/\tau_0^2$$

increasing rapidly for $\tau \gg \tau_0$ i.e. in the saturation region. For $\tau \lesssim \tau_0/2$ the linearity

$$\tau \simeq \tau_0 H_z/H_{z\infty} = \sqrt{2} H_z |\tan\psi - \tan\delta| \cos\psi/u \qquad (13)$$

holds to better than 11%. As can be seen from (12) and Fig. 3, by varying the tilting angle the time scale constant τ_0 can be made both larger and smaller than the value

$$\tau_0(\delta = 0) = H' \tan\psi/u \qquad (14)$$

corresponding to the untilted case [1]. The experimental results of Wyatt and Marinero [9], and Saltiel et al. [11] correspond to the case of large τ_0 i.e. negative δ. Choosing positive tilting angles, especially $\delta \simeq \psi$, very small τ_0's can be obtained, i.e. the femtosecond region becomes accessible for pulse duration measurements. Indeed, in the untilted case for femtosecond pulses (10b) or (13) yield a small H_z which is completely overruled by divergency, diffraction and walk-off effects, whereas setting $0 < \delta \simeq \psi$ results in a blowed-up H_z. In the latter case, however, the precise knowledge of δ becomes increasingly important as the limit $\delta = \psi$, corresponding to $\tau_0 = 0$ is approached.

It should be pointed out that the approximation of slowly varying amplitudes and (10b) break down in the case of the pulse consisting only of a small number of oscillations. The suggested experimental method may be applicable even in this case. For this, however, a solution of the second-order wave equation becomes necessary in order to replace (10b) by a more valid $H_z = H_z(\tau)$ function.

More serious problems have to be faced due to the spread of femtosecond wave packets. The spread in the nonlinear crystal can be reduced by using very thin crystals, even at a price of intensity loss which should not be crucial for femtosecond pulses. The spread during the "tilting" step, however, can be very important. In the case of a grating the spread is accompanied also by a divergence of the diffracted beam. In the case of a Michelson echelon the problems may be reduced by choosing a material of especially low-dispersion near the wavelength used and by minimizing the size of the echelon along the optical path.

In summary, we have presented an extension of the SHB method down to the femtosecond region, where a new limit due to pulse shape distortion may be expected.

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