

# Analysis of the SHG effects for characterizing ultrashort pulses with SPIDER

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The analyses of second harmonic generation (SHG) effects have been firstly conducted for characterizing ultrashort pulse with the spectral phase interferometry for direct electrical reconstruction (SPIDER). The results show that we should multiply a modulation function for the recorded interferometric intensity in order to avoid the effect of bandwidth of the pulses, thus we can exactly reconstruct the fundamental-pulse intensity. The root-mean-square (RMS) phase error generated by bandwidth is proportional to the nonlinear-crystal length, the intersection angle of the beams. We can also obtain greater phase-matching acceptance angle in type II phase-matching crystal.

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Over the last two decades, remarkable progress has been achieved in short-pulse generation directly from lasers, the shortest pulse is less than 5 fs, even as short as single-cycle pulse regime<sup>[1]</sup>. Meantime, convenient and exact measurement technology should be developed for the generation and application of ultrashort pulse. Spectral phase interferometry for direct electrical reconstruction (SPIDER) uniquely combines several advantageous elements found in pulse measurement devices. SPIDER requires no moving parts and uses a direct, noniterative retrieval algorithm that produces an unambiguous phase and intensity profile for the measured pulse. In addition, SPIDER has been shown to be accurate<sup>[2]</sup>, fast and capable of measuring pulses generated from oscillation or chirped pulses<sup>[3,4]</sup>.

Since SPIDER was invented in 1998, several studies has addressed the performance of SPIDER. SPIDER can measure the pulse as short as 3.4 fs<sup>[5]</sup>, but the phase match is inaccurate. In this paper, we analyze the second harmonic generation (SHG)<sup>[6]</sup> and its effects for characterizing ultrashort pulses, and derive a detailed description of SHG SPIDER performance for ultrabroad-band pulse with type II noncollinear phase matching.

Core of SPIDER's theoretical underpinnings is spectral shearing interferometry<sup>[7]</sup>. First, we assume that the appropriate crystal thickness should mostly be adopted from the phase-matching conditions. As it has been pointed out<sup>[8]</sup>, pulse broadening due to crystal bulk dispersion is negligibly small compared to the group-velocity mismatch for theory of SHG in a nonlinear crystal. We assume that the second harmonic (SH) field is not absorbed in the nonlinear crystal. This is well justified for any pulse. Absorption bands of the crystals are transparent in the visible start at 200 nm for Gaussian pulse. Consequently, at these frequencies the field amplitude decreases by a factor  $\exp(-\pi^2/2 \ln 2)$  compared to its maximum at 400 nm. We also assume the efficiency of SHG to be low enough to avoid depletion of the fundamental beams.

We assume that wavefronts of the fundamental waves inside the crystal are practically flat. Therefore, we treat the SHG as a function of the longitudinal coordinate only and include the transversal coordinates at the last step to account for the spatial beam profile. In fact, we should

use a convex lens or concave mirror to avoid the beam divergence, that is to say, pulses are measured with non-collinear phase-matching model. The geometry is shown in Fig. 1.

Now we can obtain the equation of wave propagation in the nonmagnetic, loss-free and homogeneous medium from Maxwell's equations<sup>[9]</sup>

$$\frac{\partial^2 E_s(z, t)}{\partial z^2} = \mu_o \frac{\partial^2 D(z, t)}{\partial t^2} + \mu_o \frac{\partial^2 P^{(2)}(z, t)}{\partial t^2}, \quad (1)$$

where  $E_s(z, t)$  is the SH field,  $P^{(2)}(z, t)$  is nonlinear polarization.  $D(z, t) = \epsilon_o \epsilon(t) E_s(z, t)$ ,  $\epsilon(t)$  is the relative permittivity. In the frequency domain, an equivalent of Eq. (1) is

$$\frac{\partial^2 \tilde{E}_s(z, \Omega)}{\partial z^2} + k_s^2(\Omega) \tilde{E}_s(z, \Omega) = -\mu_o \Omega^2 \tilde{P}^{(2)}(z, \Omega), \quad (2)$$

where  $\tilde{E}_s(z, \Omega)$  and  $\tilde{P}^{(2)}(z, \Omega)$  are Fourier transforms of  $E_s(z, t)$  and  $P^{(2)}(z, t)$ ,  $\Omega$  is the SH frequency, and  $k_s(\Omega)$  is the wavevector of the SH field,  $k_s^2(\Omega) = \Omega^2 \mu_o \epsilon_o \tilde{\epsilon}(\Omega)$  with  $\tilde{\epsilon}(\Omega)$  being the Fourier transform of the relative permittivity  $\epsilon(t)$ .

In order to simplify the Eq. (2), we write the SH field as a plane wave propagating along the  $z$  axis:

$$\tilde{E}_s(z, \Omega) = \tilde{\epsilon}_s(z, \Omega) \exp[ik_s(\Omega)z]. \quad (3)$$

Hence Eq. (2) becomes

$$\frac{\partial^2 \tilde{\epsilon}_s(z, \Omega)}{\partial z^2} + 2ik_s(\Omega) \frac{\partial \tilde{\epsilon}_s(z, \Omega)}{\partial z} = -\mu_o \Omega^2 \tilde{P}^{(2)}(z, \Omega) \exp[-ik_s(\Omega)z]. \quad (4)$$

Now we should calculate the second-order polarization  $\tilde{P}^{(2)}(z, \Omega)$  to obtain the SH field. We assume that two fundamental waves cross in the  $xz$  plane at a small angle  $2\alpha$  (see Fig. 1). The inclination with the  $z$  axis of each beam inside the crystal is then  $\alpha(\omega) = \arcsin[\sin \alpha_o/n(\omega)] \approx \alpha_o/n(\omega)$ . The delay for off-axis components of the beam due to the geometry can be expressed for a plane wave as:  $\tau \approx Ln(\omega) \tan \alpha(\omega)/2c \approx L\alpha_o/2c$  for the beam propagating in  $+\alpha$  direction. The

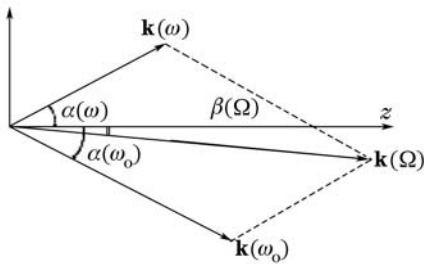


Fig. 1. Noncollinear phase matching for three wave interaction.

electric fields in the frequency domain can be found via Fourier transforms:

$$\begin{aligned}\tilde{E}_1(\omega) &= \tilde{\varepsilon}(\omega) \exp[i\omega(L\alpha_o/2c)], \\ \tilde{E}_2(\omega_o) &= \tilde{\varepsilon}(\omega_o) \exp[i\omega_o(-L\alpha_o/2c)].\end{aligned}\quad (5)$$

With SPIDER for characterizing ultrashort pulse, the second-order dielectric polarization is induced at frequency by a quasi-monochromatic fundamental pulse and a ultrabroad-band pulse, the  $\tilde{P}^{(2)}(z, \Omega)$  can be calculate as

$$\begin{aligned}\tilde{P}^{(2)}(z, \Omega) &= \varepsilon_o \tilde{\chi}^{(2)}(\omega, \omega, \omega_o) \tilde{E}_1(\omega) \tilde{E}_2(\omega_o) \\ &= \varepsilon_o \exp(i\Omega L\alpha_o/2c) \tilde{\chi}^{(2)}(\omega, \omega, \omega_o) \tilde{\varepsilon}(\omega) \tilde{\varepsilon}(\omega_o) \\ &\quad \times \exp\{i[k_z(\omega)z + k_z(\omega_o)z - \omega_o L\alpha_o/c]\},\end{aligned}\quad (6)$$

where  $\tilde{\chi}^{(2)}(\omega, \omega, \omega_o)$  is the nonlinear susceptibility. Consequently, we apply the slow-varying amplitude approximation<sup>[9]</sup>, Eq. (4) can be readily solved by integration over the crystal length  $L$ . The electric of the SH therefore becomes

$$\begin{aligned}\tilde{\varepsilon}_s(L, \Omega) &= \frac{i\Omega^2 L}{2k(\Omega)c^2} \exp(i\Omega L\alpha_o/2c) \tilde{\chi}^{(2)}(\omega, \omega, \omega_o) \\ &\quad \times \tilde{\varepsilon}(\omega) \tilde{\varepsilon}(\omega_o) \exp\{i[\Delta k(\omega, \omega_o)L/2 - \omega_o L\alpha_o/c]\} \\ &\quad \times \text{sinc}[\Delta k(\omega, \omega_o)L/2],\end{aligned}\quad (7)$$

where  $\Delta k(\omega, \omega_o)$  is the phase mismatch, which is given for type II phase matching by

$$\begin{aligned}\Delta k(\omega, \omega_o) &= K_o \cos(\alpha_o n_o(\omega)) + K_e \cos(\alpha_o n_e(\omega_o)) \\ &\quad - K_e \cos(\beta(\omega, \omega_o)) \\ &\approx K_o(\omega) + K_e(\omega_e) - K_e(\Omega).\end{aligned}\quad (8)$$

So far, we have conducted the SH field generated by nonlinear crystal for SPIDER. Due to one of the fundamental pulses is a quasi-monochromatic pulse, the group-velocity mismatch between fundamental pulse and SH is sharply decreased, and the SH field is much more simple compared to the general SHG. The geometrical smearing of the delay is studied by Taft *et al.*<sup>[10]</sup>. We can analyze this issue analogously and ignore the effect of smearing.

In this section, our goal is to obtain the effects of several parameters by SHG for SPIDER. First, we should make some approximations in order to simplify Eq. (7). For a classical harmonic-oscillator model<sup>[11]</sup>, we estimate dispersion of the second-order susceptibility  $\tilde{\chi}^{(2)}(\Omega, \omega, \omega_o)$  from the dispersion of the refractive index

$$\tilde{\chi}^{(2)}(\Omega, \omega, \omega_o) \propto \tilde{\chi}^{(1)}(\Omega) \tilde{\chi}^{(1)}(\omega) \tilde{\chi}^{(1)}(\omega_o),\quad (9)$$

where  $\tilde{\chi}^{(1)}(\Omega) = n^2(\Omega) - 1$ , Eq. (7) becomes

$$\tilde{\varepsilon}_s(\Omega) \propto F(\Omega) \tilde{\varepsilon}(\omega),\quad (10)$$

where

$$\begin{aligned}F(\Omega) &= \frac{i\Omega L}{2n_e(\Omega)c} [n_e^2(\Omega) - 1][n_o^2(\omega) - 1][n_e^2(\omega_o) - 1] \\ &\quad \times \tilde{\varepsilon}(\omega_o) \text{sinc}[\Delta k(\omega, \omega_o)L/2] \exp(i\Omega L\alpha_o/2c) \\ &\quad \times \exp\{i[\Delta k(\omega, \omega_o)L/2 - \omega_o L\alpha_o/c]\} \tilde{\varepsilon}(\omega_o).\end{aligned}\quad (11)$$

According to the theory of SPIDER, we should pay attention to the ac portion of the interferogram

$$\begin{aligned}D^{(+ac)}(\omega) &\propto F^2(\Omega) |\tilde{\varepsilon}(\omega) \tilde{\varepsilon}(\omega - \Delta\Omega)| \\ &\quad \times \exp\{-i[\Delta\phi + \phi_\omega(\omega) - \phi_\omega(\omega - \Delta\Omega)]\},\end{aligned}\quad (12)$$

where the spectral shear  $\Delta\Omega$  is

$$\begin{aligned}\Delta\phi &= [\Delta k(\omega, \omega_o) - \Delta k(\omega - \Delta\Omega, \omega_o + \Delta\Omega) \\ &\quad + 2\Delta\Omega\alpha_o/c]L/2.\end{aligned}\quad (13)$$

As shown in Eqs. (11) and (12), we can see that the fundamental-pulse intensity is not proportional to the intensity of SH. The relation between them is the factor  $F^2(\Omega)$ . We define  $F^2(\Omega)$  as modulation function, the graph of the function is shown in Fig. 2. While we measure intensity of the fundamental pulse, one of the delayed test pair should be converted with the stretched pulse. So, the SH field should be multiply the factor  $1/|F(\Omega)|^2$  in order to obtain the exact fundamental-pulse intensity. Also, we consider the sinc function, the result is that SPIDER is insensitive to the phase mismatch because of ultrabroad band. SPIDER is much more accurate than other technique for characterizing sub-10 fs optical pulse.

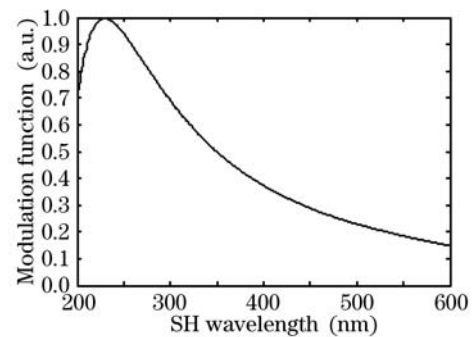


Fig. 2. Modulation function of SH field.

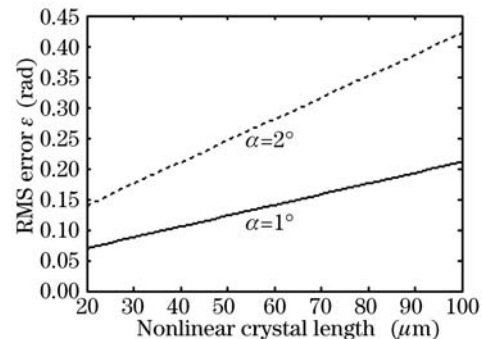


Fig. 3. Phase error for the nonlinear crystal length.

The phase difference from the interferogram of SH will add phase error that induced by noncollinear SHG. The value of the additional phase can be estimated from Eq. (13). The spectral shear is less than 1% of the bandwidth, Eq. (13) can be simplified as

$$\Delta k(\omega, \omega_o) - \Delta k(\omega - \Delta\Omega, \omega_o) \ll \Delta\Omega\alpha_o/c. \quad (14)$$

Now we can analyze the effects generated by intersection angle of the beams and the nonlinear crystal length, that is to say, we obtain the phase error due to the broadband of the input pulse. It is useful to use a root-mean-square(RMS) error<sup>[12]</sup> to define the phase error. The phase error depended on crystal length and intersection angle as shown in Fig. 3, while the input pulse has a duration of 20 fs and the value of the spectral shear is  $2\pi \times 0.9$  THz. The result is that RMS error is proportional to the nonlinear crystal length and the intersection angle. The phase error and RMS error that we estimated numerically correspond to the value that some papers addressed<sup>[13,14]</sup>. We should adopt less intersection angle and less length of the crystal from this conclusion in the practical instrument.

As mentioned above, we had assumed that wavefronts of the fundamental pulse was flat. In fact, the divergent laser beams have finite angular spreads. According to Fourier optics, an ideal flat wave can be regarded as combination of lots of flat waves. We can analyze the phase mismatch induced by beam divergence with type II phase matching to calculate phase-matching acceptance angle.

The SH spectral intensity will decrease by a factor  $\text{sinc}^2[\Delta k(\omega, \omega_o)L/2]$  due to beam divergence. Now, we expand  $\Delta k$  into Taylor series around  $\theta_m$ :

$$\Delta k = \frac{\omega \sin 2\theta_m \Delta\theta}{2c} \{ [n_e^\omega(\theta_m)]^3 [(n_e^\omega)^{-2} - (n_o^\omega)^{-2}] - 2[n_e^{2\omega}(\theta_m)]^3 [(n_e^{2\omega})^{-2} - (n_o^{2\omega})^{-2}] \}. \quad (15)$$

where  $\theta_m$  is the phase-matching angle. When  $\Delta k = \pi/L$ , we gain that the value of the phase matching acceptance angle is  $1.5 \times 10^{-2}$  rad for a pulse with wavelength at 800 nm and a 200- $\mu\text{m}$  nonlinear crystal. The angle in type II phase matching is much more greater than that in type I phase matching, that is to say, the efficiency of frequency doubling increases in SPIDER.

SPIDER is a powerful and accurate pulse diagnostic technique that is ideally suited for the measurement of a

vast variety of pulses. In this paper, we have developed the SHG theory and provided the modulation function for the intensity of fundamental pulse. We have also successfully estimated the additional phase error derived from crystal length and intersection angle and calculated the phase-matching acceptance angle. The well-developed theory makes it possible to design more accurate apparatus.

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