

Direct measurement of synchronization between femtosecond laser pulses and a 3 GHz radio frequency electric field inside a resonant cavity

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We demonstrate a method to measure synchronization between femtosecond laser pulses and the electric field inside a resonant 3 GHz radio frequency (RF) cavity. The method utilizes the Pockels effect in a crystal inside the RF cavity by measuring the retardation of the components of polarization as a function of RF phase. Resolution of the setup used is shown to be 29 ± 2 fs (root-mean-square, rms), with timing jitter between the laser pulses and the RF field inside the cavity of 96 ± 7 fs (rms). The method provides a tool to reduce jitter and improve time-resolution in ultrafast electron diffraction experiments. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4823590>]

Synchronization between femtosecond laser pulses and an oscillator operating in the radio frequency (RF) range is crucial in several experiments. In high energy accelerators and XFEL (X-ray Free Electron Laser), where photo-emission is used to generate electron bunches and RF fields to accelerate them, the energy, bunch duration, and arrival time depend on the phase of the RF field at the time of photo-emission.^{1–3} In ultrafast electron diffraction measurements, RF cavities are used to compress electron bunches below 100 fs.^{4–7} Gao *et al.*⁸ and Chatelain *et al.*⁹ have used ponderomotive scattering to determine the jitter in arrival time between compressed electron bunches and femtosecond laser pulses. They find that the temporal Instrument Response Function (IRF), the convolution between timing jitter, laser pulse, and electron bunch duration, is of the order 300–500 fs full width half maximum (fwhm), or 150–200 fs root mean square (rms) for bunches with 0.1–0.6 pC charge. Recently, Gao *et al.* have performed single shot measurements using a laser-triggered streak-camera and found arrival time jitter of 200 fs (rms). This is, at least partly, attributed to the synchronization of the RF field with respect to the laser pulses that generate the electrons and excite the sample under investigation. Different synchronization schemes have been developed and compared. Best performance has been shown with a phase-locked-loop/voltage-controlled-oscillator (PLL-VCO), where an RF oscillator is controlled using a higher harmonic of the laser oscillator.¹⁰ However, no direct measurements of the phase of the electric field in the cavity have been made. The jitter in the phase of the electric field is related to the timing jitter between the laser and electron pulses and provides an independent diagnostic. In this letter, we present results from a method that measures the phase of the electric field inside a resonant RF cavity. The method uses electro-optical sampling of the birefringence induced in a potassium dideuterated phosphide (DKDP) crystal by measuring the retardation of one of the components of the

polarization with respect to the other.¹¹ The time resolution of the method is determined as well as the timing jitter of one of the most commonly used PLL-VCO synchronization systems. When combined with an accelerator or compression cavity, the method may be used as a single-shot phase monitor in ultrafast electron diffraction experiments.

In order to measure the retardation of a femtosecond laser pulse induced by the electric field of the 3 GHz RF, we designed a resonant pillbox cavity with a DKDP crystal^{12,13} inside the cavity, see Figure 1. The crystal (EKSMA Optics) has a diameter of 9 mm and a length of 20 mm. The cavity was designed using CST Microwave Studio¹⁴ with an inner diameter of 12 mm to make it resonant in the TM_{010} mode at 3 GHz. A tuning plunger in the side of the cavity allows tuning of the resonant frequency over a range of 100 MHz.

The 3 GHz source is a PLL-VCO built by AccTec BV, based on previous work by Kiewiet *et al.*¹⁵ The femtosecond laser oscillator (FemtoLasers GmbH Femtosource) operates at 75 MHz, which is measured by a fast photodiode (Centronic AEPX65). In the PLL-VCO, the 3 GHz VCO signal is divided by 8 and locked to the 5th harmonic of the laser repetition frequency at 375 MHz. The 3 GHz is amplified to 200 W (peak power) using a power amplifier (Microwave Amps Ltd. AM83) and fed into the cavity. An external trigger pulse sets the pulse duration of the 3 GHz output of the PLL-VCO. In these experiments, the output pulse duration was set to 3 μ s with a repetition frequency of 30 Hz.

The same laser oscillator that provides the timing signal at 75 MHz is used as the probe to measure the electric field inside the DKDP crystal. The laser pulses (3.5 nJ per pulse) pass through a separate Pockels cell that is activated with 10 ns pulses with a repetition frequency of 30 Hz to reduce the repetition frequency of the laser pulses to the same frequency at which the amplifier delivers the 3 GHz pulses to the cavity. The change in polarization of the laser pulses induced by the electric field in the RF cavity is measured in a balanced diode setup for optical biasing, similar to those used in electro-optical sampling experiments,^{16–18} see Figure 2.

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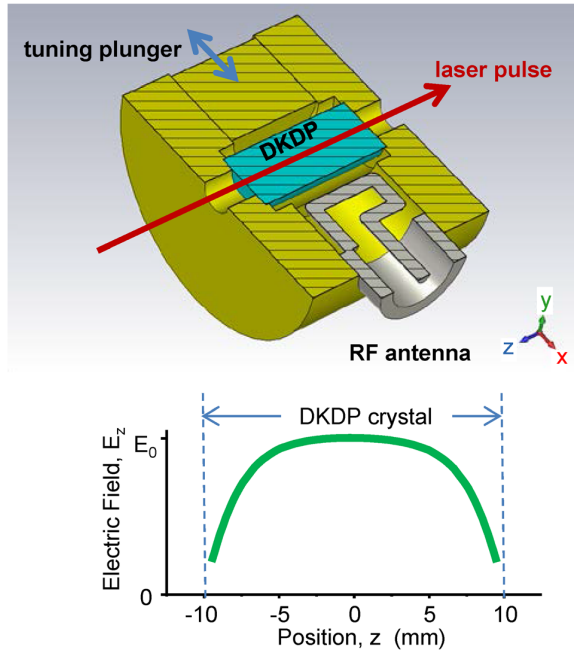


FIG. 1. RF cavity with DKDP crystal inside. The cavity was designed using CST Microwave Studio to be resonant at 3 GHz in the TM_{010} mode. The laser pulses pass through the crystal along the z -axis. The graph shows the simulated electric field distribution along the z -axis.

The linearly polarized femtosecond laser pulses pass through a quarter wave plate to make the pulses circularly polarized. After passing through the cavity, the vertical and horizontal polarization components of the pulses are split in a Wollaston prism and detected by two photodiodes. Additional electronics amplify the difference between the two signals by a factor of 21. The amplified difference and the sum of the two signals as well as each signal separately are measured using a 12-bit ADC.

The DKDP crystal is placed inside the RF cavity with its extraordinary axis along the z -axis. The center of the cavity is at $z = 0$. The electric field on the z -axis, E_z , is given by

$$E_z(z, t) = E_0(z) \cos(2\pi f_{RF} t) \quad (1)$$

with E_0 the maximum field on-axis and $f_{RF} = 3$ GHz the RF frequency. The change in retardation, Γ , between the components of the polarization is given by

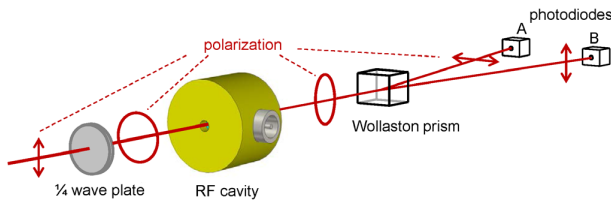


FIG. 2. Setup for electro-optic sampling of the electric field in the DKDP crystal inside the RF cavity. A linearly polarized laser pulse passes through a $1/4$ wave plate to make it circularly polarized. The principal axes of the crystal inside the cavity have been rotated by 45° with respect to the initial polarization. When the laser pulse experiences a net electric field, while passing through the crystal, the polarization becomes elliptical. The Wollaston prism splits the two components of the polarization, which are detected by a balanced photodiode setup.

$$\frac{d\Gamma(E_z)}{dz} = \frac{2\pi}{\lambda} (n_a(E_z) - n_b(E_z)) \quad (2)$$

with n_a and n_b the refractive indices along the principal axes. For the DKDP crystal, Eq. (2) reduces to

$$\frac{d\Gamma(E_z)}{dz} = 2\pi \frac{r_{63} n_0^3}{\lambda} E_z(z) \quad (3)$$

with r_{63} the longitudinal Pockels coefficient, λ the wavelength of the laser pulse in vacuum, and $n_0 = 1.49$ the refractive index in of the crystal in the absence of an electric field. The electric field is time-dependent, following Eq. (1). Since the laser pulse duration is much shorter than the period of the RF signal, the electric field at the position z^* of the laser pulse can be written as

$$E_z(z^*, \varphi_0) = E_0(z^*) \cos\left(2\pi f_{RF} \frac{n_0}{c} z^* + \varphi_0\right) \quad (4)$$

with φ_0 the phase of the RF at the moment, the laser pulse passes the center of the cavity. The phase retardation of the laser pulse is thus given by

$$\Gamma(\varphi_0) = \int_{-l/2}^{l/2} 2\pi \frac{r_{63} n_0^3}{\lambda} E_0(z^*) \cos\left(2\pi f_{RF} \frac{n_0}{c} z^* + \varphi_0\right) dz^* \quad (5)$$

with $l = 20$ mm the length of the crystal. For an electric field that is constant along the z -axis, the retardation is

$$\Gamma(\varphi_0) = \frac{r_{63} n_0^3}{\lambda} E_0 \frac{2c}{n_0 f_{RF}} \sin\left(\pi f_{RF} \frac{n_0 l}{c}\right) \cos(\varphi_0). \quad (6)$$

This equation is plotted in Figure 3. The inset in Figure 1 shows the simulated field distribution along the z -axis. When the deviation from a constant field is taken into account in the integration with the relevant values for f_{RF} and n_0 , Eq. (6) decreases by 17%. This can be accounted for by using an effective length $l_{eff} = 16.5$ mm instead of the actual length of the crystal. However, the eventual measurement is a relative measurement so that the exact values of the constants in Eq. (6) play no role in the determination of the jitter.

Equation (6) shows that the phase retardation is maximal at $\varphi_0 = 0$. The sensitivity to changes in the RF phase is maximal at $\varphi_0 = (\pm)\pi/2$, where the total phase retardation is 0,

$$\frac{\partial \Gamma(\varphi_0)}{\partial \varphi_0} = \frac{2c r_{63} n_0^2}{\lambda f_{RF}} E_0 \sin\left(\pi f_{RF} \frac{n_0 l}{c}\right) \sin(\varphi_0). \quad (7)$$

For small variations of the phase around $\varphi_0 = \pi/2$, this can be converted to a time-delay, $\Delta\tau = \Delta\varphi_0 / 2\pi f_{RF}$, of the RF-phase with respect to the moment, the femtosecond laser pulse passes through the center of the cavity, so that

$$\Delta\Gamma = \frac{4\pi c r_{63} n_0^2}{\lambda} E_0 \sin\left(\pi f_{RF} \frac{n_0 l}{c}\right) \Delta\tau \quad (8)$$

or

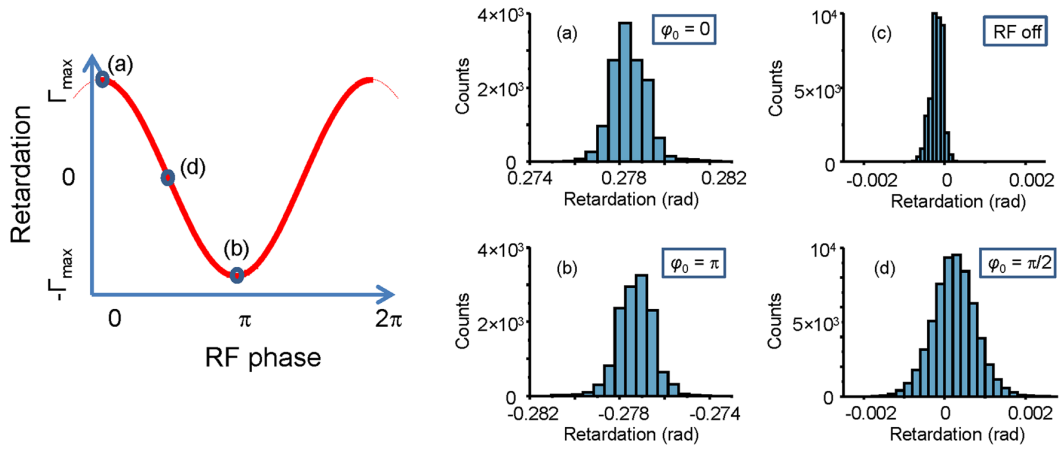


FIG. 3. The figure on the left shows how the retardation between the polarization directions of the laser pulse changes with the timing between the laser pulse and the 3 GHz RF phase. Measurements were taken at (a) $\varphi_0 = 0$ and (b) $\varphi_0 = \pi$ to determine Γ_{\max} . (c) Shows the noise of the setup with RF power switched off. (d) Is the distribution of the retardation at $\varphi_0 = \pi/2$, where the sensitivity to timing jitter is greatest.

$$\Delta\tau = \frac{1}{2\pi f_{RF}} \frac{\Delta\Gamma}{\Gamma_{\max}} \quad (9)$$

with Γ_{\max} the value of Γ at $\varphi_0 = 0$. All parameters on the right hand side of Eq. (9) are determined experimentally, so that precise knowledge of the crystal properties and the cavity is not required.

The two diodes, A and B in Fig. 2, measure the vertical and horizontal polarization components of the laser pulse. The difference is divided by the sum of the two signals to correct for fluctuations in laser power. With the principal axes of the crystal at 45° with respect to the x- and y-axes of Figure 1, the retardation can now be determined from the measured signals

$$\sin(\Gamma) = \frac{A - B}{A + B}. \quad (10)$$

First, Γ_{\max} in Eq. (9) was determined by setting the RF phase to, respectively, 0 and π . Histograms of these measurements, taken at 30 Hz sampling frequency are shown in Figs. 3(a) and 3(b). The normalized difference between the two diode signals was found to be 0.2743 with rms noise of 8×10^{-4} , corresponding to retardation, Γ_{\max} , of 277.9 mrad, with rms noise of 0.8 mrad. We then determined the resolution of the setup by switching off the RF power. The rms noise of this measurement was equivalent to $\sigma_{P_{RF}=0} = 0.15 \pm 0.01$ mrad (see Fig. 3(c)). For the uncertainty in $\sigma_{P_{RF}=0}$, we have used the standard deviation of 40 subsets of 1000 points each. The apparently skewed distribution of Fig. 3(c) is a numerical artifact, caused by the binning procedure, because the variation in magnitude of the signals is close to the resolution of the 12-bit ADC.

Combining $\sigma_{P_{RF}=0}$ with Eq. (9) and the measurement of Γ_{\max} , the rms time resolution of the setup is found to be 29 ± 2 fs. With the RF power switched on and the phase of the RF signal set to $\pi/2$, the histogram shown in Fig. 3(d) is obtained with $\sigma_{\varphi_0=\pi/2} = 0.52 \pm 0.03$ mrad. This value is a combination of timing jitter and measurement noise (as determined from the resolution measurement). The rms retardation caused by timing jitter is found:

$\sigma_{\text{timing}} = \sqrt{\sigma_{\varphi_0=\pi/2}^2 - \sigma_{P_{RF}=0}^2} = 0.50 \pm 0.04$ mrad. This corresponds to rms timing jitter between the laser and the RF field inside the cavity of 96 ± 7 fs. The data of Fig. 3(d) are shown in Fig. 4 as a function of time.

The experiments show that it is possible to measure the timing jitter between a femtosecond laser pulse and the phase of the 3 GHz RF field inside a resonant cavity with a resolution of 29 fs. The synchronization system, in combination with the laser oscillator and RF amplifier, results in a timing jitter of less than 100 fs. This is worse than earlier results of Kiewiet *et al.*,¹⁵ where we found that a similar synchronization system locks the 3 GHz to a 375 MHz oscillator with jitter of less than 20 fs. However, these results are not inconsistent, because the PLL-VCO is only one possible source of jitter. The photodiode that provides the 75 MHz to the system can heat up locally, even with the relatively low laser energy (a few pJ) used for the timing signal, resulting in a variable time delay between the arrival of the laser pulse on the photodiode and the signal from the photodiode to the PLL-VCO. Also, noise in the laser oscillator, e.g., due to mode competition in the pump laser, may contribute to the measured timing jitter. On the other side of the PLL-VCO, the RF amplifier may add to the measured jitter. The width of the distribution of Γ_{\max} , $\sigma_{\varphi_0=0}$, is actually a direct

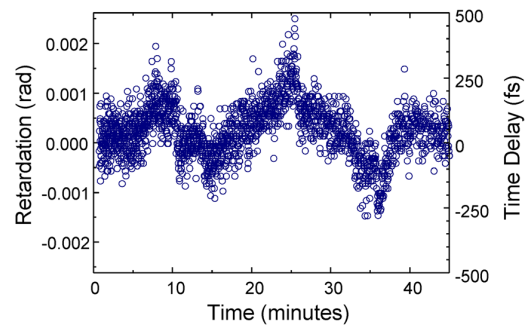


FIG. 4. Single shot measurements of the retardation at $\varphi_0 = \pi/2$. These data correspond to Fig. 3(d). For clarity, only every 30th shot (one shot per second) is shown in this figure. The axis on the right gives the relative time delay between the laser pulse and the RF phase, using Eq. (9).

measurement of the fluctuations of the electric field strength in the crystal and therefore of the power fluctuations of the amplifier. Timing jitter does not contribute to these measurements since $\partial\Gamma(\varphi_0)/\partial\varphi_0$ is 0 at $\varphi_0=0$. The rms noise of 0.8 mrad in the measurements of Γ_{\max} corresponds to relative fluctuations of the electric field strength in the crystal of 3×10^{-3} . Vice versa, these power fluctuations do not contribute directly to the measured timing jitter, because the total (integrated) field strength is 0 when $\varphi_0 = \pi/2$. However, power fluctuations, temperature changes, and electronic noise inside the amplifier may affect the phase of the amplified RF signal. The method demonstrated in this paper to measure the overall jitter between the laser pulse and the phase of the 3 GHz provides an excellent tool to track down and, where possible, eliminate these sources of jitter.

The measured timing jitter of 100 fs is a confirmation of the result by Van Oudheusden,⁶ where we found <100 fs using a synchronized streak cavity, and in line with the findings of Gliserin.¹⁰ Gao^{7,8} and Chatelain⁹ measured the jitter in arrival time for bunches that are compressed using an RF compression cavity. Pasmans¹⁹ has shown that at the position of the time focus, where the electron bunches are fully compressed, the jitter in arrival time is equal to the RF phase jitter. Gao and Chatelain found arrival time jitter of 150–200 fs (rms) in similar setups using the same synchronization system (AccTec BV) as used in the experiments here. Their results are comparable to what we measured here, considering their setups contain additional elements (electron beam line and optics) that may account for the difference.

The timing resolution of the experiments described here was shown to be 29 fs. This is in part limited by the accuracy with which we can measure the change in polarization, i.e., 0.15 mrad. This is already quite good compared to other electro-optical sampling experiments using a balanced diode configuration,¹⁸ considering that we cannot use averaging or lock-in techniques in this experiment. Still, the noise in these measurements is dominated by noise in the A/D conversion and may be improved upon. On the other side, the resolution in terms of femtoseconds is directly proportional to the maximum achievable retardation. This is ultimately limited by electrical breakdown in the RF cavity, which we have seen occasionally when using the full 200 W RF power. An improved design of the antenna that couples the RF power into the cavity, in combination with a more powerful amplifier can improve the resolution. With these improvements, 10 fs timing resolution seems feasible.

The measurements in this letter are essentially single-shot measurements of the time delay between the laser pulse and a chosen RF phase. A compression cavity is now being

developed with a DKDP crystal incorporated in it. With such a cavity, measurement of the polarization change of the probe pulse may be used for time-sorting of the shots. We have demonstrated here that the RF phase can be measured with a resolution of 0.15 mrad. If the phase of the RF field is recorded in each shot, even if the jitter is of the order of 100 fs, it may become possible to sort the arrival time of such bunches with a resolution of only a few femtoseconds.

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