

## Multi-cycle THz pulse generation in poled lithium niobate crystals

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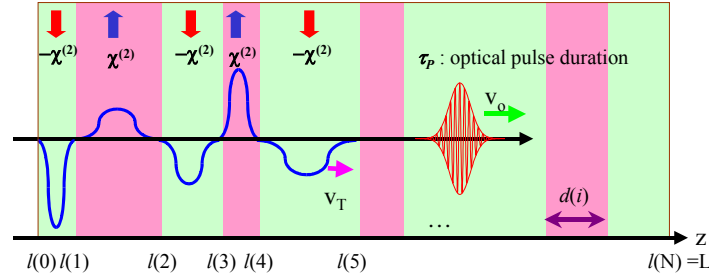
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The terahertz (THz) frequency range (0.1~10 THz) lies between the microwave and infrared regimes of the electromagnetic (EM) spectrum, but THz science and technology lags considerably behind the microwave and infrared regimes due to significant limitations in coherent THz generation and detection. The highest frequency achieved by electronic technologies does not exceed a few hundred GHz. Infra-red optical devices typically generate and detect EM waves at tens of THz at best, although a few far-infrared gas lasers and very recently quantum cascade lasers have provided coherent cw radiation at select frequencies in the THz. Recently THz technology has been an extremely active field of research, and the development of new THz sources and detectors has been filling the “THz gap.” These new technologies have great potential to integrate the electronic and optical devices, which is expected to enable ultrahigh-speed computation and communications beyond signal switching rates of 100 Gigabits/sec. Also, several research groups are exploring the potential of THz time-domain imaging for diverse applications such as package inspection and medical diagnosis, and time-domain molecular spectroscopy of rotational and vibrational dynamics is one of the promising applications of THz technology for identification and characterization of molecules.

One approach to THz generation is via nonlinear optics with ultrafast lasers: since the few-THz frequency range corresponds to the inverse of femtosecond pulse duration, femtosecond lasers have been widely used to generate ultra-broadband, single-cycle THz waves. On the other extreme, free-electron lasers and quantum cascade lasers have been used as continuous-wave (cw) THz sources. Many applications, however, will require arbitrary THz waveform generation, as opposed to single-cycle or cw THz sources, in a similar fashion to the user-specified waveforms used in optical pulse shaping or the RF waveforms used in NMR. The first step toward flexible THz waveform generators has been achieved recently: it has been demonstrated that complex multi-cycle THz pulses can be generated via optical rectification in poled lithium niobate (PLN) crystals using femtosecond optical pulses.

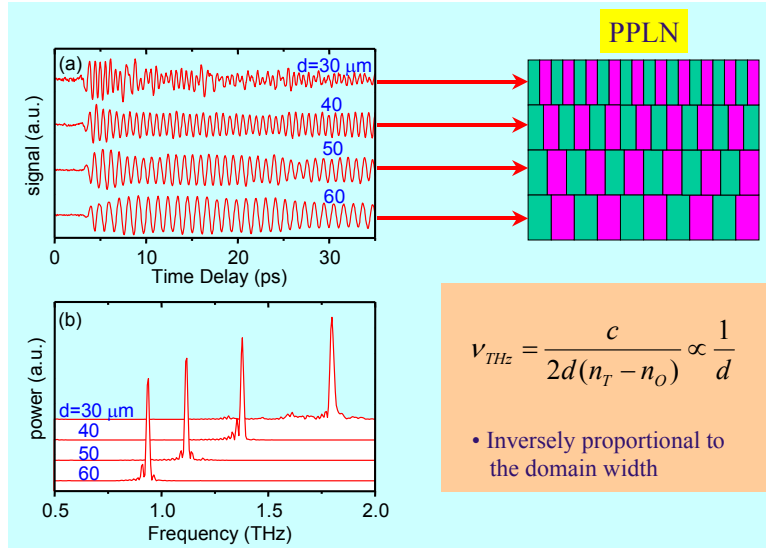
The scheme of the multi-cycle THz waveform generation is shown in Fig. 1, which illustrates optical rectification in the pre-engineered domain structure of a PLN crystal. The second order nonlinear susceptibility ( $\chi^{(2)}$ ) of the crystal reverses sign between neighboring domains. The green (down) and purple (up) shades in Fig. 1 indicate the direction of the optic axis. When a femtosecond optical pulse propagates through a PLN crystal with such a domain structure, a THz nonlinear polarization is generated via optical rectification. Due to the mismatch between the optical group velocity and the THz phase velocity (e.g., the optical and THz indices of refraction of  $\text{LiNbO}_3$  are  $n_O = 2.3$  and  $n_T =$

5.2 respectively), the optical pulse will lead the THz pulse by the optical pulse duration  $\tau_p$  after a walk-off length  $l_w = c\tau_p/(n_T - n_O)$ . If the domain length ( $d(i)$ ) of the poled nonlinear crystal is comparable to the walk-off length, each domain in the crystal contributes a half cycle to the radiated THz field. Since the length of the half-cycle pulse is proportional to the corresponding domain length, the resulting THz pulse directly corresponds to the crystal domain structure as illustrated in Fig. 1.



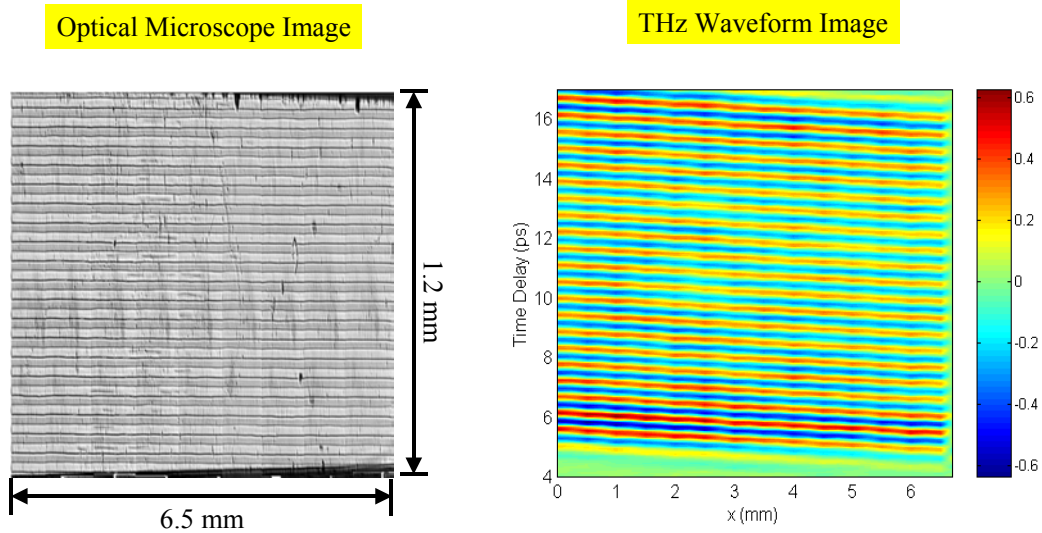
**Figure 1. Schematic diagram of the multi-cycle THz pulse generation in a PLN crystal. The green (down) and purple (up) shade indicates the direction of the domain optic axis.**

When the domain structure is periodic, i.e., the domain lengths are identical ( $d(i) = d$ ), narrow-band multi-cycle THz pulses can be generated. Figure 2 shows the THz waveforms and the respective spectra from a periodically-poled lithium niobate (PPLN) crystal when the domain width is 30, 40, 50, and 60  $\mu\text{m}$  at  $T = 115 \text{ K}$ .<sup>1</sup> The optical pump spectrum is centered at 800 nm and the pulse duration is 150 fsec. The generated THz waves are focused by a pair of off-axis paraboloidal mirrors into a 1 mm (110)-cut ZnTe crystal. The THz waveform is measured by free-space electro-optic sampling.<sup>2</sup> The sample is a z-cut PPLN crystal, which is laterally chirped, i.e., multiple domain structures of slightly different domain width were fabricated side by side at a regular distance from one to another. The domain width varies from 30 to 60  $\mu\text{m}$  with 5  $\mu\text{m}$  step size. The relative bandwidth  $\Delta\nu/\nu$  of the THz field is given simply by  $2/N$  in the absence of absorption and domain-width fluctuations, where  $N$  is the number of domains in the PPLN. The frequency of the THz wave is determined as  $\nu_{\text{THz}} = c/2d(n_T - n_O)$  where  $c$  is the light velocity,  $d$  is the domain-width, and  $n_T(n_O)$  is the group refractive index at THz (optical) frequency. Tuning of the THz frequency is accomplished simply by scanning the sample laterally to the beam propagation direction. Figure 2 clearly shows that the THz wave from the shorter domains is generated with higher frequency as expected. Continuous THz frequency tuning also has been demonstrated using fanned-out PPLN crystals in which the domain width varies continuously across the lateral direction.



**Figure 2. (a) THz waveforms and (b) power spectra at  $T = 115$  K when the PPLN domain-width is 30, 40, 50, and 60  $\mu\text{m}$ . The sample is a laterally chirped z-cut PPLN crystal: multiple domain structures of slightly different domain width are fabricated side by side at a regular distance from one to another. The THz frequency is inversely proportional to the domain width.**

It is interesting to note that, since the THz waveform is essentially a direct manifestation of the crystal domain structure, the multi-cycle THz generation method provides a novel approach to characterizing the domain structure of poled materials. Since the domain structure of the materials affects the physical properties of the crystals in many aspects, it is important to understand the patterns of the domain structure. One direct method is the observation of differentially etched domains using surface probing tools, although this results in the inevitable destruction of the sample. On the other hand, probing the inner crystal domain structure by analyzing the terahertz (THz) waveforms generated by optical rectification in ferroelectric crystals should be a nondestructive technique. Figure 3(a) shows an optical image of the electrode pattern of a PPLN crystal, magnified to exaggerate the domain lengths.<sup>3</sup> The optic axis of the sample is normal to the surface. It is clearly seen that the sample is wedged. The domain structure of the sample is supposed to match with the electrode pattern, but in reality each domain suffers width fluctuations due to the strong coercive electric fields and field leakage out of the electrodes. To probe the entire domain structure of the PPLN, the PPLN sample was scanned laterally with respect to the fixed excitation beam. The optical excitation beam is vertically positioned in the middle of the sample. The THz waveform image of the PPLN domain structure is shown in Fig. 3(b), which indicates that the domain structure is fairly periodic, which is consistent with the electrode pattern.<sup>3</sup> Fifteen data points are taken along the x-axis; values between the data points are interpolated. In the narrower region of the crystal, the effective optical beam path is shorter, thus the THz waves arrive earlier than in the wider region. The slanted domain interfaces to the sample surfaces are also well reproduced in the THz waveform image.



**Figure 3. (a) Optical microscope image of electrode patterns (magnified in the direction of 1.2 mm) and (b) two dimensional image of THz waveforms of a 6.5 mm  $\times$  1.2 mm PPLN crystal (THz field amplitude corresponds to the color-bar).**

Arbitrary THz waveforms can be obtained using non-periodic PLN crystals. Figure 4 shows experimental results and numerical simulations for three different types of PLN structures, which demonstrates the feasibility and versatility of the THz pulse shaping scheme.<sup>4</sup> First, the zero-area double pulse (Fig. 4(a)) consisting of two pulses with a  $\pi$  phase shift is generated from a domain structure in which a single domain (100- $\mu\text{m}$ ) is placed between two sets of multiple domains (50  $\mu\text{m}$ ). The corresponding spectrum (Fig. 4(a)) clearly shows the signature of the interference fringes of two coherent pulses. The phase-locked double pulses can be applied to coherent control experiments ranging from chemical reactions to semiconductor carrier dynamics. Second, a chirped THz waveform is shown in Fig. 4(b). The domain structure for the chirped pulse includes multiple domains ranging from 20 to 90  $\mu\text{m}$  with 1  $\mu\text{m}$  gradual increment. The broad band spectrum is shown in Fig. 4(b). Chirped pulses in the optical regime have been used in the control of atomic wavepackets via adiabatic transfer. Similarly, chirped THz pulses are applicable to adiabatic transfer between excitonic wavepackets in semiconductor nanostructures or between rotational and vibrational modes of molecules. Third, Fig. 4(c) shows the waveform from a structure of alternating domain length (30 and 60  $\mu\text{m}$ ). Narrow and broad half-cycle pulses appear alternately. Figure 4(c) shows the narrow band spectrum corresponding to the 90- $\mu\text{m}$  period. A second harmonic signal appears because of the asymmetric domain structure. It can be used to excite two spectrally separated molecular transitions simultaneously. The flexibility of this pulse shaping

technique also has been shown in a recent theoretical work demonstrating the generation of coherent narrow-band THz radiation with multi-frequency modes in an optical superlattice of ferroelectric domains arranged as a Fibonacci sequence.<sup>5</sup> In general, the results clearly show that shaped THz pulses can be generated in PLN structures. Most of the quantitative discrepancy between experiment and simulation comes from the temporal decay of the THz pulses in the experiments because of the THz absorption in LiNbO<sub>3</sub> by optical phonons. Cooling down the samples below 100 K suppresses the THz absorption significantly.<sup>1</sup>

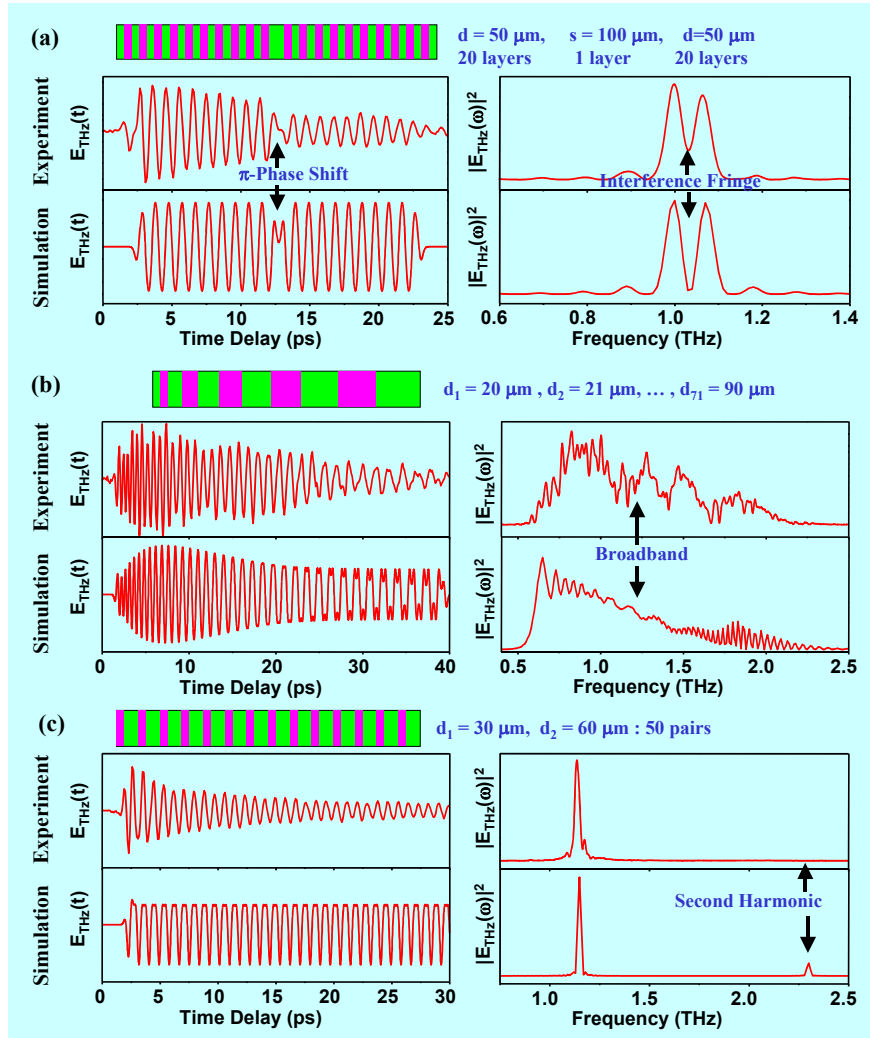


Figure 4. Experimental data and numerical solution of (a) zero-area double pulse from a domain structure in which a single domain (100- $\mu\text{m}$ ) is placed between two sets of periodic multiple domains (50- $\mu\text{m}$ ), (b) chirped pulse from a domain structure which includes multiple domains ranging from 20 to 90- $\mu\text{m}$  with 1  $\mu\text{m}$  gradual increment, and (c) pulse with alternating period from a structure of alternating domain length (30 and 60- $\mu\text{m}$ ). THz waveforms and corresponding spectra from poled lithium niobate structures are shown. Diagrams of the three domain structures are included: green (down) and purple (up) indicates the alternating direction of the crystal optic axis.

Since PPLN crystals based on quasi-phase-matching techniques have been in demand for diverse uses such as optical frequency conversion and high speed switching device for

light modulation, several companies now fabricate PPLN crystals based on customers' specifications and requirements. HC Photonics, INO, and ISOWAVE are among those companies capable of customized PLN fabrication. It is also worthwhile to refer to other crystals with different bandwidth, emission intensity, and damage threshold. Ferroelectric crystals such as  $\text{KNbO}_3$ ,  $\text{LiTaO}_3$ ,  $\text{KTiOPO}_4$  (KTP), and  $\text{RbTiOAsO}_4$  (RTA) may be used to generate multi-cycle THz pulses. Quasi-phase-matched crystals of  $\text{KNbO}_3$ , KTP, and  $\text{LiTaO}_3$  containing periodically poled structures already have been fabricated. Periodically-poled KTP crystals are commercially available from Cobolt AB. Besides ferroelectric crystals, superlattice structures of III-V semiconductors and organic crystals such as DAST might also be used in multi-cycle THz pulse generation.

Generation of arbitrary THz waveforms has great potential for applications to THz imaging systems, ultrafast optical signal processing, ultrahigh-speed computing, quantum information science, nanotechnology, and chemical reaction dynamics among other areas. THz time-domain spectroscopy permits precise measurements not only of the amplitude but also of the phase of THz waves. The phase sensitivity is vital to many applications such as high-contrast THz imaging and quantum control of semiconductor nanostructures.

#### **References:**

1. Y. -S. Lee et al., *Appl. Phys. Lett.* **78**, 3583 (2001).
2. Q. Wu and X.-C. Zhang, *Appl. Phys. Lett.* **68**, 1604 (1996).
3. Y. -S. Lee et. al., *Appl. Phys. Lett.* **77**, 2488 (2000).
4. Y. -S. Lee et. al., *Appl. Phys. Lett.* **82**, 170 (2003).
5. Y. Qin et. al., *Appl. Phys. Lett.* **83**, 1071-1073 (2003).