Tunable narrow-band terahertz generation from periodically poled lithium niobate

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We describe a technique for generating tunable narrow-band terahertz radiation via optical rectification in periodically-poled lithium niobate. Frequency tuning is accomplished by spatially chirping the domain width laterally to the beam propagation direction, and adjusting the temperature of the sample. We demonstrate tuning over a continuous range from 0.8 to 2.5 THz. The bandwidth of the terahertz waveforms is as narrow as 0.02 THz. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1373406]

There is presently considerable research activity in the development of the new sources in the terahertz (THz) region of the spectrum. The recent development of femtosecond lasers naturally brought about various techniques to generate broad-band, single-cycle THz waves, since the inverse pulse duration of femtosecond lasers corresponds to the THz frequency range. These sources have found many applications, such as THz spectroscopy for studies of carrier dynamics and intermolecular dynamics in liquids,[1,2] dielectric response of molecules, polymers and semiconductors,[3–5] etc. Since single-cycle waves intrinsically possess broad bandwidth, they are particularly useful for investigating the THz response of matter over a wide range of spectrum concurrently.

In many cases, on the other hand, spectrally narrow and bright THz sources are desired when the phenomenon under investigation has relatively narrow bandwidth. Although many ways of generating single-cycle THz waves with broad bandwidth have been developed, few schemes exist to generate narrow-band THz waves. One promising technique for generating narrow-band THz waves has been demonstrated, using optical rectification of femtosecond pulses in periodically poled lithium niobate (PPLN).[6,7] Narrow-band sources are in great demand for a wide range of applications in spectroscopy, sensing, communication, and imaging; for such applications frequency tunability is essential. In this letter, we demonstrate the frequency tuning of narrow-band THz radiation from PPLN crystals by lateral spatial chriping of the domain width and adjusting the temperature of the samples.

In the experiment, we use two types of *z*-cut PPLN crystals which were laterally chirped, i.e., multiple domain structures of slightly different domain width were fabricated side by side at a regular distance from one another. Sample 1 is 6 mm long and has seven domain structures. The domain width varies from 30 to 60 μm with a 5 μm step size. Sample 2 is 10 mm long and has 46 domain structures. The domain width varies from 21.5 to 62 μm with a 0.9 μm step size. Tuning of the THz is accomplished simply by scanning the sample laterally to the beam propagation direction; fine tuning is accomplished via temperature tuning, as will be discussed later.

Narrow-band THz wave forms are generated via optical rectification of pump pulses generated from a 250 kHz Ti:Sapphire regenerative amplifier.[8] The pump spectrum is centered at 800 nm and the pulse duration is 200 fs. Pump pulses of 50 nJ are focused to a 100 μm spot on the samples. The samples are placed in a cryostat in which the temperature of the sample can be varied continuously between 10 and 300 K. The generated THz waves are focused by a pair of off-axis parabolodial mirrors into a 1 mm (110)-cut ZnTe crystal. The THz wave form is measured by free-space electro-optic sampling.[9]

As the femtosecond optical pulses propagate through the PPLN crystal, a THz nonlinear polarization is generated via optical rectification. Each domain in the PPLN contributes a half cycle to the radiated THz field due to the walk off between the optical and THz pulses.[6,7] In the absence of absorption and domain-width fluctuations, the relative bandwidth $\Delta\nu/\nu$ of the THz field is given simply as $2/N$, where N is the number of domains in the PPLN. The frequency of the THz wave is determined as

$$\nu_T = c/2d(n_T - n_O), \tag{1}$$

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.[6] Thus, frequency tuning can be accomplished by adjusting the domain width of the PPLN. Figures 1(a) and 1(b) show the THz wave forms and the respective spectra from the sample 1 when the domain width is 30, 40, 50, and 60 μm at $T=115$ K. Figure 1 clearly shows that the THz wave from the shorter domains is generated with higher frequency as expected from Eq. (1). Some features of the THz wave forms and spectra of Fig. 1 are noteworthy. Because of the material absorption at THz frequencies, the THz wave decays as it propagates through the PPLN. The absorption is more severe at higher frequencies, because the main source of the absorption is the low-frequency tail of the transverse optical phonon mode at 7.6 THz.[10]

It has been demonstrated that the absorption of the THz waves in PPLN crystals is suppressed substantially at a low

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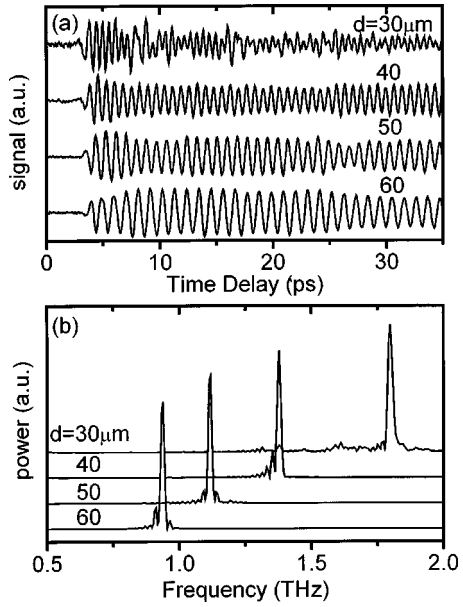

FIG. 1. (a) THz wave forms and (b) power spectra at $T=115$ K when the domain width is 30, 40, 50, and 60 μm.

temperature.[7] Figure 2 shows the measured THz wave forms and power spectra at $T=12$, 60, 90, 120, 150, 180, 210, 240, and 270 K from the 35 μm domain-width region in sample 1. The temporal decay of the THz wave form becomes important for temperatures above about 100 K, and the absorption increases gradually as the temperature increases. Due to the absorption, the bandwidth of the THz emission increases at high temperature. It is also observed that the spectrum shifts to the red with increasing temperature. The spectral shift from 12 to 270 K is 0.1 THz, which corresponds to 6.5% of the central frequency (1.55 THz at $T=150$ K) of the THz waves. The frequency shift is due to an interplay of the temperature-dependent index of refraction and the thermal expansion of the sample. Since the thermal expansion coefficient of lithium niobate is $4.1\times10^{-6}\,\mathrm{K}^{-1}$ in the normal direction of the optic axis,[11] the change of the domain width from 10 to 270 K is only 0.1%, which is negligible compared to the spectral shift. Thus, it is clear that the temperature

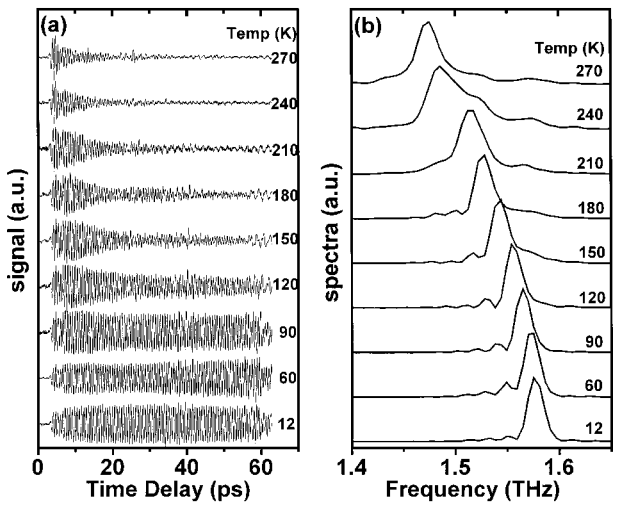

FIG. 2. Measured (a) THz wave forms and (b) spectra at $T=12$, 60, 90, 120, 150, 180, 210, 240, and 270 K from the domain structure of 35 μm domain width in the 6 mm sample.

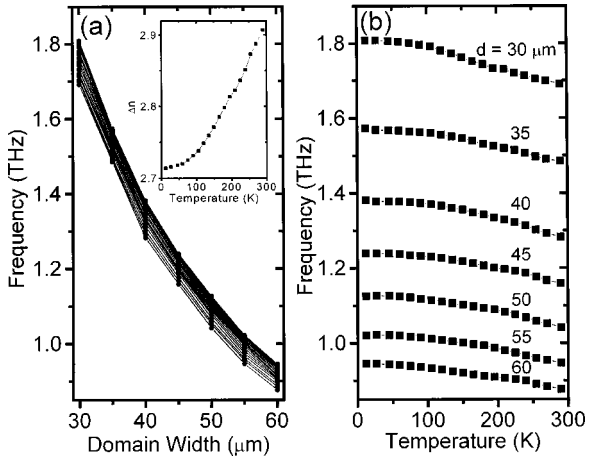

FIG. 3. (a) THz frequency vs the domain width at $T=12$, 27, 45, 60, 75, 90, 105, 120, 135, 150, 165, 180, 195, 210, 225, 240, 255, 279, and 290 K from top to bottom. Δn vs sample temperature is shown in inset. (b) THz frequency vs sample temperature for the domain width of 30, 35, 40, 45, 50, 55, and 60 μm.

dependence of the index of refraction at the THz and the optical frequencies is the dominant factor to explain the spectral shift. Temperature control can be used for the fine tuning of the frequency, though it has some disadvantage due to the wave form distortion by the temperature-dependent absorption.

Figure 3(a) shows the THz frequency versus the domain width at various sample temperatures for sample 1. The frequency is inversely proportional to the domain width for a given temperature, and shifts to the red with increasing temperature. Assuming that the thermal expansion of the sample is negligible, $\Delta n(=n_T-n_O)$ can be obtained to fit the data using Eq. (1). The fitting results are shown in the inset of Fig. 3(a), which demonstrate the increase of Δn with increasing temperature. The fitting result at room temperature ($\Delta n=2.907$) agrees well with the reported values of the optical and THz index of refraction, $n_O=2.3$ and $n_T=5.2$.[12] In Fig. 3(b), the temperature dependence of the THz frequency is shown for several domain widths. For the domain widths of 30 and 60 μm, temperature tuning covers spectral ranges of 0.14 and 0.08 THz, respectively.

Fine tuning of the frequency has been performed using sample 2. Figure 4 shows the measured (solid square) and the calculated (solid line) frequency of the THz wave forms

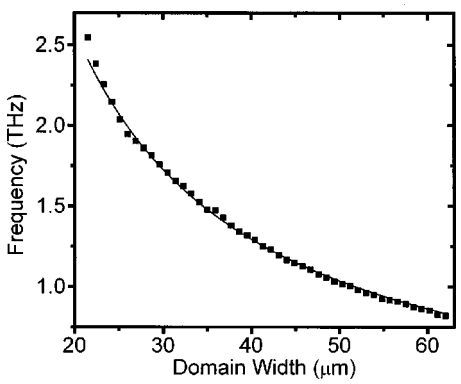

FIG. 4. Measured (solid square) and calculated (solid line) frequency of the THz waves as a function of domain width using a 10 mm z-cut PPLN at room temperature.

as a function of the domain width at room temperature. $\Delta n(=2.9)$ at room temperature has been used in the calculation of Eq. (1) to obtain the THz frequency. There is excellent agreement between the experiment and the calculation, which indicates that the THz index of refraction is nearly constant within this frequency range. The smaller step size of the domain width in sample 2 as compared to that in sample 1 results in the smaller increment of the THz frequency with decreasing domain width. The increment of the frequency with 0.9 μm step size of the sample 2 is 0.051 (0.013) THz for the domain width of 30 (60) μm. Since the temperature tuning range is much bigger than the frequency increment from the domain width change, continuous frequency tuning from 0.8 to 2.5 THz can be accomplished from sample 2 by varying the sample temperature.

In conclusion, we have demonstrated frequency tuning of narrow-band THz waves from PPLN crystals. The THz frequency can be tuned continuously from 0.8 to 2.5 THz by lateral spatial chirping of the domain widths in the PPLN crystal and adjusting the sample temperature.

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