Temperature-dependent Sellmeier equation at terahertz frequency range for 1 mol% MgO-doped stoichiometric lithium tantalate

Kyu-Sup Lee, Do-Kyeong Ko, and Nan Ei Yu

1Department of Physics and Photon Science, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea
2Advanced Photonics and Research Institute, Gwangju Institute of Science and Technology, Gwangju 61005, Republic of Korea

E-mail: neyu@gist.ac.kr

Received December 8, 2016; accepted February 9, 2017; published online March 6, 2017

We first examined a temperature-dependent Sellmeier equation for the extraordinary refractive index of MgO-doped stoichiometric LiTaO₃ (MgO:SLT) crystal via frequency-tunable narrowband terahertz (THz) generation at 0.4–1.8 THz and at a low temperature range of 80–300 K. The mean deviation of Sellmeier equation fit was less than 0.3%, being equivalent with the predicted measurement uncertainty of 0.3%. The temperature effect ($\Delta n/\Delta T$) was 2.02 x $10^{-3}$/K at 1 THz. Moreover, we showed the smaller birefringence for MgO:SLT than MgO-doped stoichiometric LiNbO₃ at 300 K from THz time-domain spectroscopy. This study is crucial for material study itself and THz device engineering.

© 2017 The Japan Society of Applied Physics

Terahertz (THz) wave spectrum lies between far-IR waves and millimeter waves corresponding to the scientific fields of photonics and electronics, respectively, which has allowed to converged both unfamiliar technologies toward the THz frequency range owing to its various benefits. Recently, THz waves have been actively researched for its applications in nondestructive imaging and spectroscopy: explosive and illegal drug detection, cancer cell diagnostics, pesticide detection, mail screening, and gas sensing. Moreover, THz waves have been highlighted as an alternative source for short-range wireless communication.

Lithium tantalate (LiTaO₃, LT) crystals have been spotlighted as high-power THz wave sources owing to their various benefits for high energy-conversion efficiency and resistance to optical damage. Also quasi-phase-matching (QPM) method, predominantly using periodically poled structures of lithium niobate (LiNbO₃, LN) and LT crystals, has been extensively used for optical frequency conversion owing to its simple and wide tunability and high conversion efficiency. Recently, QPM has been broadened to the THz frequency range via frequency down-conversion from near-IR to THz and has demonstrated the generation of multi-cycle and narrowband THz waves. By the fact that the narrowband (<100 GHz) THz waves can interacted and excited not neighboring modes but target single-mode in materials, the QPM-based THz generation scheme is crucial in high-resolution imaging and spectroscopy. Moreover, the narrowband THz waves allows to avoid the absorption band in water vapor, being useful in THz wireless communication.

However, although the information of material dispersion is crucial in designing optical devices for THz applications, the studies in the optical constants at THz frequency range have still been left behind. Particularly, only a few studies about the temperature dependence of the refractive index and absorption coefficient for LT crystal at THz range have been reported, in contrast to LN, in contrast to LN, it has been noted that LT shows better performances for high-power THz generation than LN, such as the higher resistance to optical damage, higher photorefractive damage threshold, and lower green-induced infrared absorption, which allows intense pulse pumping for significant conversion from optic to THz pulses. The larger bandgap of LT restricts the three-photon absorption at 800-nm pumping. Furthermore, the stoichiometric LT (SLT) showed a Curie temperature of 695 °C and a uniform distribution on the device wafer, which indicates that the stoichiometric method contributes not only to the significantly lower coercive field for polarization reversal but also to the lower defect density inside the wafer. The lower defect allows lower THz absorption in the material compared to congruent LT and is more suitable for the suppression of THz absorption using liquid nitrogen. For these reasons, SLT is more crucial in high-power THz generation using an intense optical pulse when compared to LN that has been most widely used as a nonlinear optical crystal for THz generation. By this, we report the temperature-dependent Sellmeier equation for SLT at THz range based on the frequency-conversion measurement that we have evaluated using a QPM-structured SLT.

We have developed the simply tunable narrowband THz generation scheme with an ultrashort optical pulse. THz waves are generated via difference-frequency generation (DFG) between two arbitrary frequency components in a femtosecond (fs) pulse of a pump laser. By wave-vector conservation rule in DFG process, the generated THz frequency is as follows:

$$v_{\text{THz}} = \frac{c}{(n_{\text{THz}} \pm n_{\text{opt}})\Lambda},$$

where $c$ is the speed of light in vacuum, $n_{\text{THz}}$ and $n_{\text{opt}}$ are refractive indices for THz and optical waves, respectively, and $\Lambda$ is a QPM period. The DFG process is naturally determined by the QPM conditions for THz generation in the forward ("-" sign) and backward ("+" sign) directions. As the generated THz frequency is determined by QPM condition, the fan-out QPM structure with linearly increased QPM period simply allows to tune the generated THz frequency. Consequently, the THz refractive index can be inferred by Eq. (1) from the varying THz frequencies under various QPM periods and temperatures.

In the experiment, we used the same THz generation and detection geometry shown in Ref. 26. A fs optical pulse

Content from this work may be used under the terms of the Creative Commons Attribution 4.0 license. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Based on a Ti:sapphire oscillator system with a pulse duration of 190 fs, a wavelength of 800 nm, and a bandwidth of 9 nm was used at a repetition rate of 76 MHz. The optical pump beam with a 450-mW power was focused onto a THz source crystal with a Gaussian waist of about 50 µm. The generated THz pulse was detected by a low-temperature grown GaAs photoconductive antenna with a dipole gap of 5 µm. We fabricated a fan-out, periodically poled 1 mol% MgO-doped SLT (PPMgO:SLT) with a thickness of 1.5 mm and a length of 10 mm, as shown in Fig. 1. The QPM period linearly varied from 50 to 90 µm along the width of 40 mm. The sample was cooled down from room temperature (300 K) to the liquid nitrogen temperature of 80 K.

First, the modulation of a multi-cycle THz pulse at a QPM period of 90 µm was evaluated in the time domain under various temperatures. Figure 2(a) shows the recovery of THz electric field at lower temperatures as a result of the lower THz absorption caused by the coupling between transverse optical phonons of the crystal and the polaritons of input infrared waves. The temperature dependence of the generated THz waveforms from the other QPM periods had the same characteristics of that from 90 µm with different frequencies.

Figures 2(b) and 2(c) show the corresponding THz center frequency modulation under several QPM periods and temperatures. The results indicated the blue shift in the THz frequency at both shorter QPM periods and lower temperatures. Therefore, the continuous tuning of center frequency from 0.85 to 1.65 THz at the fan-out PPMgO:SLT structure was obtained under temperatures between 80 and 300 K, as shown in Fig. 2(b). The corresponding narrower spectral bandwidth at lower temperature was obtained, which exhibited the narrowest bandwidth of up to 17 GHz at 0.85 THz and the temperature of 80 K. The bandwidth is predicted by the following equation \( \Delta \nu_{THz} = 2\nu_{THz}/N \), where \( N \) is the number of QPM domains along the interaction length between optical and THz pulses. At a 75-µm QPM period, the domain number was 133 and the predicted bandwidth was 15.8 GHz, which was consistent with the experimental value of 17 GHz within the error in domain numbers caused from the fabrication. The broader spectra and the lower intensities were obtained at higher temperatures, which implies the lower accuracy of the measurement of a THz center frequency at high temperature. However, the measurement at the highest temperature of 300 K could give a good accuracy of the measurement because the bandwidth was less than 0.1 THz and could give the much better accuracy of center frequency than 0.1 THz corresponding to smaller error than 0.5%.

Moreover, we considered the thermal contraction of a QPM period at lower temperatures as follows:

\[
\Lambda(T) = \Lambda_0(T_0)[1 + \alpha(T - T_0) + \beta(T - T_0)^2],
\]

where \( \alpha \) and \( \beta \) are the linear and quadratic thermal expansion coefficients of 1.45762 × 10^{-5} K^{-1} and 2.68608 × 10^{-8} K^{-2}, respectively, and \( \Lambda_0 \) is the QPM period at room temperature (300 K, \( T_0 \)). From Eq. (2), we obtained small thermal shrinkage in actual QPM periods, e.g., a 90-µm period at 300 K was shortened to 89.7 µm at 80 K and the maximum contraction ratio was less than 0.1%.
We compared the refractive indices for several kinds of LT that have been specified by other researchers at room temperature (300 K). References 15, 30, and 31 provided the refractive indices for congruent LT (CLT) in the form of dielectric function from the infrared reflectivity measurement, far-infrared absorption spectroscopy, and THz time-domain spectroscopy (THz-TDS), respectively, and those are plotted in dash-dot curves, as shown in Fig. 3. Recently, Tokodi et al.25 reported the refractive index for 1 mol % Mg-doped SLT (Mg:SLT) via THz-TDS, being plotted as a dash curve in Fig. 3. The difference of the refractive indices for LTs at room temperature was obtained because the compared samples were slightly different kinds of the LT crystals in terms of its dopant existence and doping ratio, which induces the variations in crystal composition of [Li] and [Ta]. Different manufacturers can give the slight composition variations as well, which induced the difference of indices for CLTs from Refs. 15, 30, and 31, as plotted in dash-dot curves in Fig. 3. Meanwhile, the dispersion of LT with the consideration of the temperature effect was only given by this report in the form of Sellmeier equation. We revealed the temperature dependence of refractive index at 1 THz as \(dn_{THz}/dT = 2.02 \times 10^{-3}/K\) from the derived Sellmeier equation. Moreover, MgO:SLT presented the smaller refractive index than CLT and Mg:SLT, which implies the smaller dispersion \((\Delta n = n_{THz} - n_{opt})\) over THz and optical frequency ranges. Thus, MgO:SLT can exhibit the best performance in the high-power THz generation owing to its low Fresnel loss at the surfaces and small walk-off effect between optical and THz pulses as well.

In addition, we compared the refractive indices for SLT and stoichiometric LN (SLN) at a THz range 0.1–2 THz via a commercial THz-TDS system (TeraView TPS 3000) in the frequency range of 0.6–2 THz with a spectral resolution of 0.0075 THz (0.25 cm\(^{-1}\)) at room temperature (300 K). Both \(\gamma\)-cut 1 mol % MgO-doped SLT and SLN (MgO:SLN) crystals were used to measure the refractive indices for both ordinary and extraordinary waves. By rotating the crystal by an angle of 90°, we easily changed the direction of the optical axis of the crystals according to the transmitted polarization of a THz pulse in THz-TDS so that the refractive indices along both ordinary and extraordinary axes could be measured. By careful measurement in THz-TDS system, the good accuracy of the refractive index was obtained even though the MgO:SLT crystals used in the narrowband THz generation method and the THz-TDS were not the same which could induce the slight variations in crystals compositions and the following slight variations in refractive index. In the result, we obtained the discrepancy of \(n_{THz}\) less than 1% of its absolute value, which was valid range in obtaining \(n_{THz}\) for MgO:SLT crystals. Figure 4(a) shows the larger refractive indices for MgO:SLT than for MgO:SLN at both axes and the larger indices along ordinary axis \((n_0)\) than along extraordinary.

### Table I. Temperature-dependent Sellmeier coefficients.

| Parameter | Value | Parameter | Value |
|-----------|-------|-----------|-------|
| \(a_0\)   | 36.9  | \(b_0\)   | 6.36 \times 10^{-3} |
| \(a_1\)   | 53200 | \(b_1\)   | 0.115 |
| \(a_2\)   | 125   | \(b_2\)   | 7.16 \times 10^{-4} |

We used the Sellmeier equation to describe that the material dispersion has taken the temperature effects into consideration, which was a simplified version of the equation presented by Abedin et al.29 in terms of the THz frequency as

\[
\frac{n^2(\nu)}{\nu^2} = (a_0 + b_0f) + \frac{(a_1 + b_1f)\nu^2}{c^2 - (a_2 + b_2f)^2\nu^2},
\]

where \(a_0, a_1, a_2, b_0, b_1,\) and \(b_2\) are the Sellmeier coefficients, \(f\) is the temperature factor given by \(f = (T - 25.5^\circ C)/(T + 570.82^\circ C)\), \(c\) is the speed of light in vacuum in \(\mu m/ps\), \(\nu\) is the frequency in THz, and \(T\) is the temperature in \(^\circ C\). The refractive index values at each temperature were fitted by Eq. (3), and the parameters were determined via the least-squares optimization. The averaged deviation was smaller than 0.3% of its absolute values, which was consistent with the uncertainties of 0.3% with which the refractive indices were calculated. The fitted refractive indices at several temperatures are shown as solid curves in Fig. 3, and the corresponding Sellmeier coefficients are listed in Table I.

![Fig. 3.](image-url) (Color online) Extraordinary refractive indices for three types of LT crystals: 1 mol % MgO:SLT from our measurement (scatter and solid curves) under several temperatures, Mg:SLT from Ref. 25 (dashed curve and blue) and CLTs from Refs. 15, 30, and 31 (dash-dot curves and pink, orange, and green, respectively) at room temperature (300 K).
MgO-doped SLT and SLN. extraordinary waves and (b) the corresponding birefringence for 1 mol %

Jpn. J. Appl. Phys. showing the lower birefringence for MgO:SLT than for MgO:SLN, as shown in Fig. 4(b), and it was consistent
cated the same characteristics for LT and LN given by Schall et al.31) The MgO:SLT showed smaller birefringence than the MgO:SLN, as shown in Fig. 4(b), and it was consistent
tendency for LT and LN.31)

In summary, we present the temperature-dependent Sellmeier equation for MgO:SLT crystal at a frequency range of 0.4–1.8 THz under a low temperature range of 80–300 K. Refractive indices along extraordinary axis were obtained from THz-TDS, showing the lower birefringence for MgO:SLT than for MgO:SLN. The investigated temperature-dependence and birefringence are essential for design and engineering of optical devices.

Acknowledgments This research was supported by National Research Foundation of Korea grants funded by the Ministry of Education, Science and Technology (2013-1A1A3007880 and NRF-2015M2A2A4A03044653), by the “Ultrashort Quantum Beam Facility Program” through a grant provided by Gwangju Institute of Science and Technology, and by the “Public Institution Linked Regional Industry Development Program” funded by the Ministry of Trade, Industry, and Energy in Korea.

1) A. G. Davies, A. D. Burnett, W. Fan, E. H. Linfield, and J. E. Canningham, Mater. Today 11 [3], 18 (2008).
2) R. M. Woodward, V. P. Wallace, R. J. Pye, B. E. Cole, D. D. Armone, E. H. Linfield, and M. Pepper, J. Invest. Dermatol. 120, 72 (2003).
3) Y. Hua and H. Zhang, IEEE Trans. Microwave Theory Tech. 58, 2064 (2010).
4) H. Hoshina, Y. Sasaki, A. Hayashi, C. Otsa, and K. Kawase, Appl. Spectrosc. 63, 81 (2009).
5) D. M. Mittleman, R. H. Jacobsen, R. Neelamani, R. G. Bari, and M. C. Nuss, Appl. Phys. B 67, 379 (1998).
6) T. Nagatsuma, S. Horiguchi, Y. Minakata, Y. Yoshimizu, S. Hisatake, S. Kuwano, N. Yoshimoto, J. Terada, and H. Takahashi, Opt. Express 21, 23736 (2013).
7) A. Chernykh, V. Shur, E. Nikolaeva, E. Shishkin, A. Shur, K. Terabe, S. Kurimura, K. Kitamura, and K. Gallo, Mater. Sci. Eng. B 120, 109 (2005).
8) R. L. Byer, J. Nonlinear Opt. Phys. Mater. 6, 549 (1997).
9) Y.-S. Lee, T. Meade, V. Perlín, H. Winful, T. B. Norris, and A. Galván, Appl. Phys. Lett. 76, 2505 (2000).
10) N. E. Yu, C. Kang, H. K. Yoo, C. Jung, Y.-L. Lee, C.-S. Kee, D.-K. Ko, J. Lee, K. Kitamura, and S. Takekawa, Opt. Photonics News 15 [3], 34 (2004).
11) S. Matsuura, M. Tani, H. Abe, K. Sakai, H. Ozeki, and S. Saito, J. Mol. Spectrosc. 187, 97 (1998).
12) J. Federici and L. Moullet, J. Appl. Phys. 107, 111101 (2010).
13) W. D. Johnston and I. P. Kaminov, Phys. Rev. 168, 1045 (1968).
14) A. S. Barker, A. A. Ballman, and J. A. Ditzenberger, Phys. Rev. B 2, 4233 (1970).
15) L. Pálffalvi, J. Hebling, J. Kuhl, Á. Péter, and K. Polgár, J. Appl. Phys. 97, 123505 (2005).
16) R. Sorade, I. Breunig, C. Tulea, and K. Buse, Appl. Phys. B 99, 63 (2010).
17) J. Kiessling, K. Buse, and I. Breunig, J. Opt. Soc. Am. B 30, 950 (2013).
18) X. Wu, C. Zhou, W. R. Huang, F. Ah, and F. X. Kártner, Opt. Express 23, 29729 (2015).
19) Y. Furukawa, M. Nakamura, S. Takekawa, K. Kitamura, T. Hatanaka, K. Nakamura, H. Ito, A. Alexandrovski, and M. M. Fejer, in Advanced Solid-State Lasers, ed. C. Marshall (Optical Society of America, Washington, D.C., 2001) OSA Trends in Optics and Photonics Series, Vol. 50, p. 658.
20) N. E. Yu, S. Kurimura, Y. Nomura, M. Nakamura, K. Kitamura, Y. Takada, J. Sakuma, and T. Sumiyoshi, Appl. Phys. Lett. 85, 5134 (2004).
21) D. A. Scrymgeour and V. Gopalan, Phys. Rev. B 71, 184110 (2005).
22) K. Kitamura, Y. Furukawa, K. Niwa, V. Gopalan, and T. E. Mitchell, Appl. Phys. Lett. 73, 3073 (1998).
23) N. E. Yu, K. S. Lee, D.-K. Ko, C. Kang, S. Takekawa, and K. Kitamura, Opt. Commun. 284, 1395 (2011).
24) L. Tokodi, A. Buzády, J. Hebling, and L. Pálffalvi, Appl. Phys. B 122, 235 (2016).
25) N. E. Yu, M.-K. Oh, H. Kang, C. Jung, B. H. Kim, K.-S. Lee, D.-K. Ko, S. Takekawa, and K. Kitamura, Appl. Phys. Express 7, 012101 (2014).
26) N. S. Stoyanov, D. W. Ward, H. Feurer, and K. A. Nelson, Nature 41, 95 (2002).
27) I. Dolev, A. Ganany-Padowicz, O. Gayer, A. Arie, J. Mchangama, and G. Gift, Appl. Phys. B 96, 423 (2009).
28) K. S. Abedin and H. Ito, J. Appl. Phys. 80, 6561 (1996).
29) K. P. Cheung and D. H. Axt, Infrared Phys. 26, 23 (1986).
30) M. Schall, H. Helm, and S. R. Keiding, Int. J. Infrared Millimeter Waves 20, 595 (1999).

Fig. 4. (Color online) (a) Refractive indices along ordinary and extraordinary waves and (b) the corresponding birefringence for 1 mol % MgO-doped SLT and SLN.