Widely linear and non-phase-matched optical-to-terahertz conversion on GaSe:Te crystals

Wei-Chen Chu,[1] Shin An Ku,[1] Harn Jiunn Wang,[1] Chih Wei Luo,[1,*] Yu. M. Andreev,[2] Grigory Lanskii,[2] and T. Kobayashi[1,3]

[1]*Department of Electrophysics, National Chiao-Tung University, Hsinchu, Taiwan*

[2]*Institute of Monitoring of Climatic and Ecological Systems of Siberian Branch of Russian Academy of Sciences, Tomsk, Russia*

[3]*Advanced Ultrafast Laser Research Center, and Department of Engineering Science, Faculty of Informatics and Engineering, The University of Electro-Communications, Chofugaoka 1-5-1, Chofu, Tokyo 182-8585, Japan*

**Corresponding author: cwluo@mail.nctu.edu.tw*

Received October 17, 2011; revised January 11, 2012; accepted January 12, 2012; posted January 13, 2012 (Doc. ID 156558); published February 28, 2012

We demonstrate the widely linear and broadband terahertz (THz) generation on GaSe:Te crystals by femtosecond laser pulses. It was found that the dopant, Te atoms, in GaSe crystals significantly enhances the efficiency of THz generation, and its central frequency can be tuned by varying the crystal thickness through non-phase-matched optical rectification. Moreover, the wide-ranging linearity for the optical-to-THz conversion and central-frequency-tunable THz generation promise for GaSe:Te crystals to be potential materials for high-power (>1.36 μW) THz applications. © 2012 Optical Society of America

OCIS codes: 190.4400, 320.7130.

Despite that terahertz (THz) waves [1] have been developed only in the past few decades, a large number of useful applications of the THz wave have been developed, such as the measurements of molecular vibrational modes and time-resolved THz spectroscopy, which have led to a new "tera era." Therefore, for practical applications, the development of novel sources and methods in the THz range has become a key issue. A relatively compact and economical avenue for generating coherent THz radiation is taking advantage of optical conversion in nonlinear crystals (e.g., GaSe, $LiNbO_3$, GaP, GaAs, and $ZnGeP_2$) [2–6]. Among these nonlinear crystals, layered ε-GaSe crystals possess a number of advantages, including a large nonlinear coefficient and significant birefringence, which have been widely used in far-IR generation [3] and mid-IR spectroscopy [7]. Nevertheless, for further application in optics, improvement in optical and mechanical properties of GaSe crystals by doping proper elements is highly desired. High-quality GaSe:Te crystals have been prepared because Te atoms slightly strengthen the mechanical properties [8] and modify the optical and electrical properties noticeably [9,10]. However, GaSe:Te crystals have not yet been studied in THz generation.

This study reports THz wave generation from GaSe:Te crystals by femtosecond (fs) laser pulses and shows that THz generation efficiency was improved substantially. Compared to the commonly used ZnTe crystals, GaSe:Te crystals can provide widely linear optical-to-THz conversion with an optical pumping fluence of up to 6.9 mJ/cm^2.

The p-type pure and Te-doped GaSe single crystals grown by the Bridgman method [11] were used in this study. The concentrations of Te in all GaSe:Te crystals were determined by electron probe microanalysis (with an accuracy of 0.01 wt. %) are 0.01, 0.07, 0.38, 0.67, and 2.07 mass %, respectively. On the other hand, a commercial Ti:sapphire oscillator operating at a central wavelength of 800 nm and with pulse duration of 100 fs with a repetition rate of 5.2 MHz was used as a pumping source. Under the scheme of normal incidence (the polarization is along the a–b plane), the pump beam with 3.5 mJ/cm^2 and diameter of 46 μm was focused on z-cut GaSe:Te crystals to generate THz waves. Any residual 800 nm laser beam was blocked by a Si wafer. The transmitted THz pulses were collected and focused on a 100 μm thick (110)-oriented ZnTe crystal by gold-coated off-axis parabolic mirrors. The electro-optical (EO) sampling technique was subsequently applied to detect the emitted THz fields in the time domain. All experiments were performed in a dry nitrogen-purged box.

Typical THz waveforms at various azimuthal angles φ were generated from a 0.38 mass % GaSe:Te crystal, as shown in Fig. 1(a). Here, φ is defined as the angle between the [100] direction of (001) GaSe:Te crystals and the polarization of the incident optical pulse. Considering EO detection and the $\bar{6}2m$ point group of the crystals, the electric field of THz generated from GaSe:Te crystals by optical rectification can be derived as

$$E_{\mathrm{THz}} \propto \chi_{22} \cos 2\theta \sin 3\varphi, \tag{1}$$

where θ is the angle between the c axis of a crystal and the direction of the incident light. The right inset in Fig. 1(a) shows the intensity of the main peak in THz signals as a function of φ, which can be well described by the φ-dependent relation of Eq. (1). Therefore, the THz generation mechanism on GaSe:Te crystals is optical rectification, which is different from the difference-frequency mixing in $ZnGeP_2$ [6]. The sixfold symmetry of THz intensity is attributed to the hexagonal structure on the (001) plane of GaSe:Te crystals. Moreover, similar sixfold symmetry observed for all GaSe:Te crystals

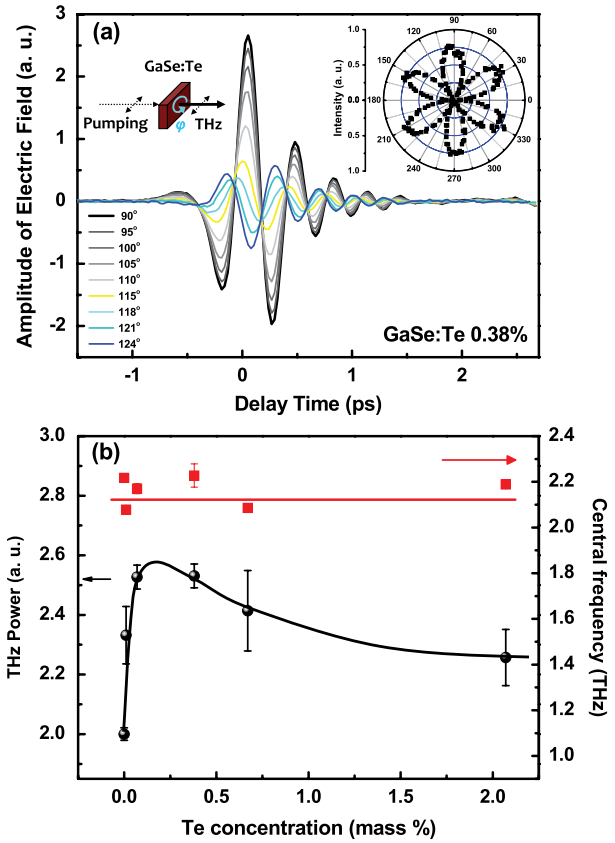

Fig. 1. (Color online) (a) Temporal waveforms of THz radiation at various azimuthal angle φ on a 0.38 mass % GaSe:Te crystal with thickness of 0.30 mm. The right inset illustrates the intensity of the THz main peak as a function of φ. The left inset shows the configuration of THz generation on GaSe:Te crystals. (b) Power and central frequency of THz radiation on various GaSe:Te crystals with thickness of 0.30 mm.

indicates that the Te atoms doped in GaSe do not destruct the crystal structure of the GaSe matrix.

Figure 1(b) unambiguously shows the effect of Te atoms doped in GaSe crystals for THz generation. Nevertheless, the central frequencies (~2.15 THz) of THz radiation are independent of the Te concentration; we found that the THz power suddenly increases once the Te atoms are doped into the GaSe crystals, even for the small Te doping of 0.01 mass %. For 0.38 mass % GaSe:Te crystals, the output power of emitted THz is enhanced by a factor of 21% compared to the GaSe crystals. According to Vodopyanov's studies [12], the optical-to-THz conversion efficiency is proportional to $\chi^{(2)}_{ijk}$. Thus, these results imply that heavy-atom doping, that is, Te, in GaSe crystals can improve its nonlinearity. However, the optical-to-THz conversion efficiency could not increase further at higher Te-doping level because of increases in structural defects (e.g., polytypism, stacking faults, and dislocation) [9–11].

To describe the THz wave generated on GaSe:Te crystals more clearly, Fig. 2 shows a comparison of THz Fourier spectra from a GaSe crystal, a 0.38 mass % GaSe:Te crystal, and a commonly used ZnTe crystal. For GaSe and 0.38 mass % GaSe:Te crystals, the spectra of the emitted THz pulse cover the range of 0.5 ~ 5 THz. Additionally, the spectra of THz radiation from GaSe:Te crystals are

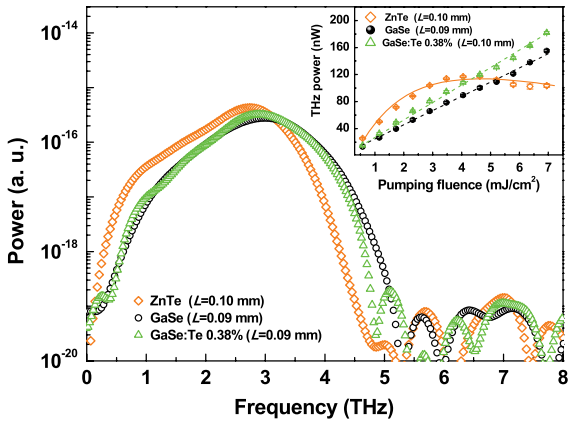

Fig. 2. (Color online) Fourier spectra of the THz radiation from ZnTe, GaSe, and 0.38 mass % GaSe:Te crystals. Inset, THz power as a function of pump fluences. L, thickness of the crystals. The solid and dashed lines are guides for the eyes.

wider than those of the ZnTe crystals with the same thickness at a high frequency side. This is caused by the frequency of the transverse optical phonon absorption peak for GaSe centered at 7.1 THz, which is higher than that of ZnTe at 5.3 THz [3,13].

The inset in Fig. 2 shows the average output power of THz radiation calibrated by a liquid He-cooled bolometer as a function of pumping fluences on ZnTe, GaSe, and GaSe:Te crystals. Because of the two-photon absorption of pumping light and the free carrier absorption of THz on ZnTe, the THz output power saturates above the pumping fluence of 3 mJ/cm^2 [14]. However, the value of the two-photon absorption coefficient of GaSe ($\beta_{\mathrm{GaSe}} = 0.558$ cm/GW) is smaller than that of ZnTe ($\beta_{\mathrm{ZnTe}} = 4.2$ cm/GW) [15], which implies that fewer free carriers are generated in GaSe:Te crystals to diminish the suppression effect in the THz output power. Therefore, the linear dependence (the originally quadratic dependence is suppressed by two-photon absorption) between the THz output power and pumping fluence is clearly observed. Consequently, at relative low pump fluences (<4.6 mJ/cm^2), the emitted THz power from GaSe:Te crystals is smaller than that from ZnTe crystals. Once the pump fluences are larger than 4.6 mJ/cm^2, however, the emitted THz power on the GaSe:Te crystals exceeds that on ZnTe crystals considerably because of the larger two-photon absorption of ZnTe. The conversion efficiency of GaSe:Te crystals was significantly higher than that of the GaSe crystal, especially in the high pumping fluence range. Thus, we can expect that GaSe:Te crystals to be a promising material for high-power THz generation, which would be attractive for applications in THz spectroscopy.

Although no significant changes are in the central frequency of THz radiation by fixing the thickness to 0.3 mm on various GaSe:Te crystals [Fig. 1(b)], the marked redshift of the central frequency is observed by changing the thickness of GaSe:Te crystals, as shown in Fig. 3. Under the slowly varying envelope approximation [12], the power spectrum of THz radiation can be described as

$$I(\omega_T, L) \propto \omega_T^2 d_{\mathrm{eff}}^2 \tau^2 L^2 \exp\left(-\frac{\tau^2 \omega_\tau^2}{4}\right) \mathrm{sinc}^2(\Delta k L/2), \quad (2)$$

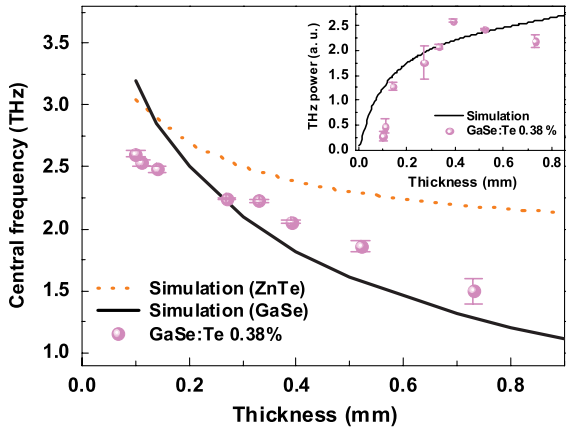

Fig. 3. (Color online) Central frequency of THz radiation on 0.38 mass % GaSe:Te crystals with various thicknesses. Inset, thickness dependence of THz power on 0.38 mass % GaSe:Te. All THz generation experiments were performed by the pumping level of 3.5 mJ/cm^2.

where ω_T is the THz angular frequency, L is the thickness of the crystal, $d_{\text{eff}} = \frac{1}{2}\chi^{(2)}$, τ is the pulse duration, and the k-vector mismatch is $\Delta k = (n_{\text{THz}} - n_{\text{opt}}^{\text{gr}})\omega_{\text{THz}}/c$. The refractive index in GaSe crystals was obtained from [16], in which $n_{\text{opt}}^{\text{gr}}(\text{at}\,800\ \text{nm}) = 3.12$ and $n_{\text{THz}}(\text{at}\,1\ \text{THz}) = 3.26$. For the different thicknesses of the crystals, L, the THz spectrum can be simulated by Eq. (2) and the central frequency of the THz radiation can be further identified from the simulated spectrum. For the thin crystals, the central frequency of THz radiation simulated from Eq. (2) is dominated by pulse duration. However, the Δk in Eq. (2) leads the change in central frequency on thicker crystals. Consequently, the central frequency of THz radiation from thin GaSe and ZnTe crystals is similar, as shown by the solid and dashed lines in Fig. 3. While the crystal thickness increases, the central frequency of THz radiation on ZnTe crystals approaches 1.9 THz, where phase matching is satisfied [17]. However, the central frequencies of THz radiation on GaSe and GaSe:Te crystals decrease gradually to 1.5 THz because of no phase matching condition being satisfied in this region. In some manner, we can consider this characteristic advantageous for tuning the THz central frequency by varying the thickness of GaSe:Te crystals. Moreover, the THz radiation power on 0.38 mass % GaSe:Te crystals increases by 8.8 times while the thickness increases from 0.1 mm to 0.52 mm [see inset of Fig. 3], which is excellently consistent with the simulation results. Therefore, the high THz radiation power on GaSe:Te crystals could be obtained simply by using thicker crystals.

In conclusion, we demonstrated broadband THz generation with widely linear optical-to-THz conversion on GaSe:Te crystals by non-phase-matched optical rectification. The dopant—Te atoms—in GaSe crystals improves the efficiency of THz generation significantly, especially in the high pumping fluence range. By increasing crystal thickness, the central frequency of THz radiation from GaSe:Te crystals shifts markedly to red, and its power increases. Furthermore, the high-power (>1.36 μW under the pumping of 6.9 mJ/cm^2 on a 0.52 mm thick 0.38 mass % GaSe:Te crystal) and central-frequency-tunable ($\Delta f_c \sim 1.5$ THz) THz radiation can be realized in GaSe:Te crystals.

This work was supported by the National Science Council of Taiwan, R.O.C. under grant nos. NSC96-2923-M-009-001-MY3 and NSC98-2112-M-009-008-MY3 and by the Grant MOE ATU Program at NCTU.

References

1. B. Ferguson and X.-C. Zhang, Nat. Mater. **1**, 26 (2002).
2. W. Shi, Y. J. Ding, N. Fernelius, and K. Vodopyanov, Opt. Lett. **27**, 1454 (2002).
3. Y.-S. Lee, T. Meade, V. Perlin, H. Winful, and T. B. Norris, Appl. Phys. Lett. **76**, 2505 (2000).
4. T. Tanabe, K. Suto, J. Nishizawa, K. Saito, and T. Kimura, Appl. Phys. Lett. **83**, 237 (2003).
5. K. L. Vodopyanov, M. M. Fejer, X. Yu, J. S. Harris, Y.-S. Lee, W. C. Hurlbut, V. G. Kozlov, D. Bliss, and C. Lynch, Appl. Phys. Lett. **89**, 141119 (2006).
6. P. Kumbhakar, T. Kobayashi, and G. C. Bhar, Appl. Opt. **43**, 3324 (2004).
7. C. W. Luo, K. Reimann, M. Woerner, T. Elsaesser, R. Hey, and K. H. Ploog, Phys. Rev. Lett. **92**, 047402 (2004).
8. A. A. Tikhomirov, Yu. M. Andreev, G. V. Lanskii, O. V. Voevodina, and S. Y. Sarkisov, Proc. SPIE **6258**, 625809 (2006).
9. S. Shigetomi and T. Ikari, J. Appl. Phys. **95**, 6480 (2004).
10. I. Evtodiev, L. Leontie, M. Caraman, M. Stamate, and E. Arama, J. Appl. Phys. **105**, 023524 (2009).
11. S. A. Ku, W.-C. Chu, C. W. Luo, Y. Andreev, G. Lanskii, A. Shaiduko, T. Izaak, V. Svetlichnyi, E. Vaytulevich, K. H. Wu, and T. Kobayashi, Opt. Express **20**, 5029 (2012).
12. K. L. Vodopyanov, Opt. Express **14**, 2263 (2006).
13. G. Gallot, J. Q. Zhang, R. W. Mcgowan, T. I. Jeon, and D. Grischkowsky, Appl. Phys. Lett. **74**, 3450 (1999).
14. M. C. Hoffmann, K.-L. Yeh, J. Hebling, and K. A. Nelson, Opt. Express **15**, 11706 (2007).
15. I. B. Zotova and Y. J. Ding, Appl. Opt. **40**, 6654 (2001).
16. C.-W. Chen, T.-T. Tang, S.-H. Lin, J. Y. Huang, C.-S. Chang, P.-K. Chung, S.-T. Yen, and C.-L. Pan, J. Opt. Soc. Am. B **26**, A58 (2009).
17. A. Nahata, A. S. Weling, and T. F. Heinz, Appl. Phys. Lett. **69**, 2321 (1996).