

Generation and spectral manipulation of coherent terahertz radiation with two-stage optical rectification

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Abstract: We propose and experimentally demonstrate the generation of single-cycle terahertz radiation with two-stage optical rectification in GaSe crystals. By adjusting the time delay between the pump pulses employed to excite the two stages, the terahertz radiation from the second GaSe crystal can constructively superpose with the terahertz field injected from the first stage. The high mutual coherence between the two terahertz radiation fields is ensured with the coherent optical rectification process and can be further used to synthesize a desired spectral profile of coherent THz radiation. The technique is also potentially useful for generating high-power single-cycle terahertz pulses, usually limited by the pulse walk-off effect of the nonlinear optical crystal used.

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References and links

1. G. M. H. Knippels, X. Yan, A. M. MacLeod, W. A. Gillespie, M. Yasumoto, D. Oepts, and A. F. G. van der Meer, "Generation and complete electric-field characterization of intense ultrashort tunable far-infrared laser pulses," *Phys. Rev. Lett.* **83**, 1578-1581 (1999).
2. G. L. Carr, M. C. Martin, W. R. McKinney, K. Jordan, G. R. Neil, and G. P. Williams, "High-power terahertz radiation from relativistic electrons," *Nature*, **420**, 153-156 (2002).
3. J. T. Darrow, X. -C. Zhang, D. H. Auston, and J. D. Morse, "Saturation properties of large-aperture photoconducting antennas," *IEEE J. Quantum Electron.* **28**, 1607-1616 (1992).
4. C. Weiss, R. Wallenstein, and R. Beigang, "Magnetic-field-enhanced generation of terahertz radiation in semiconductor surfaces," *Appl. Phys. Lett.* **77**, 4160-4162 (2000).
5. K. -L. Yeh, M. C. Hoffmann, J. Hebling, and K. A. Nelson, "Generation of 10 μ J ultrashort terahertz pulses by optical rectification," *Appl. Phys. Lett.* **90**, 171121-1-3 (2007).
6. R. Huber, A. Brodschelm, F. Tauser, and A. Leitenstorfer, "Generation and field-resolved detection of femtosecond electromagnetic pulses tunable up to 41 THz," *Appl. Phys. Lett.* **76**, 3191-3193 (2000).
7. Q. Wu and X. -C. Zhang, "Free-space electro-optics sampling of mid-infrared pulses," *Appl. Phys. Lett.* **71**, 1285-1286 (1997).
8. W. Shi and Y. J. Ding, "A monochromatic and high-power terahertz source tunable in the ranges of 2.7-38.4 and 58.2-3540 μ m for variety of potential applications," *Appl. Phys. Lett.* **84**, 1635-1637 (2004).
9. T. Bartel, P. Gaal, K. Reimann, M. Woerner, and T. Elsaesser, "Generation of single-cycle THz transients with high electric-field amplitudes," *Opt. Lett.* **30**, 2805-2807 (2005).
10. J. Dai, X. Xie, and X. -C. Zhang, "Terahertz wave amplification in gases with the excitation of femtosecond laser pulses," *Appl. Phys. Lett.* **91**, 211102-1-3 (2007).
11. X. -C. Zhang and D. H. Auston, "Optoelectronic measurement of semiconductor surfaces and interfaces with femtosecond optics," *J. Appl. Phys.* **71**, 326-338 (1992).
12. L. Xu, X. -C. Zhang, D. H. Auston, and B. Jalali, "Terahertz radiation from large aperture Si p-i-n diodes," *Appl. Phys. Lett.* **59**, 3357-3359 (1991).
13. A. Dreyhaupt, S. Winnerl, T. Dekorsy, and M. Helm, "High-intensity terahertz radiation from a microstructured large-area photoconductor," *Appl. Phys. Lett.* **86**, 121114-1-3 (2005).

14. M. Cronin-Golomb, "Cascaded nonlinear difference-frequency generation of enhanced terahertz wave production," *Opt. Lett.* **29**, 2046-2048 (2004).
15. T. Hattori and K. Takeuchi, "Simulation study on cascaded terahertz pulse generation in electro-optic crystals," *Opt. Express* **15**, 8076-8093 (2007).
16. R. M. Koehl and K. A. Nelson, "Coherent optical control over collective vibrations traveling at lightlike speeds," *J. Chem. Phys.* **114**, 1443-1446 (2001).
17. V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan, *Handbook of Nonlinear Optical Crystals* (Springer, Berlin, 1997), pp. 166-169.
18. N. B. Singh, T. Henningsen, V. Balakrishna, D. R. Suhre, N. Fernelius, F. K. Hopkins, and D. E. Zelmon, "Growth and characterization of gallium selenide crystals for far-infrared conversion applications," *J. Cryst. Growth* **163**, 398-402 (1996).
19. R. A. Kaindl, D. C. Smith, M. Joschko, M. P. Hasselbeck, M. Woerner, and T. Elsaesser, "Femtosecond infrared pulses tunable from 9 to 18 μm at an 88-MHz repetition rate," *Opt. Lett.* **23**, 861-863 (1998).
20. Q. Wu and X. -C. Zhang, "Free-space electro-optic sampling of terahertz beams," *Appl. Phys. Lett.* **67**, 3523-3525 (1995).
21. K. L. Vodopyanov, S. B. Mirov, V. G. Voevodin, and P. G. Schunemann, "Two-photon absorption in GaSe and CdGeAs₂," *Opt. Commun.* **155**, 47-50 (1998).
22. I. B. Zotova and Y. J. Ding, "Spectral measurements of two-photon absorption coefficients for CdSe and GaSe crystals," *Appl. Opt.* **40**, 6654-6658 (2001).
23. A. M. Kulibekov, K. Allakhverdiev, D. A. Guseinova, E. Yu. Salaev, and O. Baran, "Optical absorption in GaSe under high-density ultrashort laser pulses," *Opt. Commun.* **239**, 193-198 (2004).
24. M. C. Hoffmann, K. -L. Yeh, J. Hebling, and K. A. Nelson, "Efficient terahertz generation by optical rectification at 1035 nm," *Opt. Express* **15**, 11706-11713 (2007).
25. T. Löffler, T. Hahn, M. Thomson, F. Jacob, and H. G. Roskos, "Large-area electro-optic ZnTe terahertz emitters," *Opt. Express* **13**, 5353-5362 (2005).
26. F. G. Sun, W. Ji, and X. -C. Zhang, "Two-photon absorption induced saturation of THz radiation in ZnTe," in Conference on Lasers and Electro-Optics, OSA Technical Digest (Optical Society of America, Washington DC, 2000), 479-480.
27. E. Budiarto, J. Margolies, S. Jeong, J. Son, and J. Bokor, "High-intensity terahertz pulses at 1-kHz repetition rate," *IEEE J. Quantum Electron.* **32**, 1839-1846 (1996).
28. F. Blanchard, L. Razzari, H.-C. Bandulet, G. Sharma, R. Morandotti, J.-C. Kieffer, T. Ozaki, M. Reid, H. F. Tiedje, H. K. Haugen, and F. A. Hegmann, "Generation of 1.5 μJ single-cycle terahertz pulses by optical rectification from a large aperture ZnTe crystal," *Opt. Express* **15**, 13212-13220 (2007).

1. Introduction

Tremendous efforts have recently been focused on developing intense terahertz (THz) radiation sources for spectroscopy, security inspections, and biomedical imaging. Although free electron lasers and energy-recovery Linacs are able to produce intense terahertz radiation in a wide frequency range [1, 2], cost effective access to these large-scale facilities is still limited. For many applications, compact table-top terahertz radiation sources [3-13] are desirable. For example, a semiconductor biased with an electric or magnetic field and illuminated with an ultrashort laser pulse can produce an ultrafast current transient, which then radiates terahertz field efficiently [3, 4]. Terahertz radiation can also be generated by use of optical frequency mixing in nonlinear optical crystals [5-8]. Higher-order nonlinear optical processes, such as four-wave-mixing in plasma, can also be used [9, 10]. A variety of designs for THz radiation emission [11-13] have been demonstrated.

Optical rectification (OR) is one of the premier methods for generation of terahertz radiation [5-7] with advantages of broadband terahertz radiation output, high saturation fluence for the pump pulses, and scalability. Generation of near single-cycle pulses centered at 0.5 THz frequency with pulse energy up to 10 μJ was demonstrated with OR [5]. A theoretical study of cascaded difference-frequency generation (DFG) for far-infrared generation had been recently proposed [14]. The propagation length-dependent flux of generated terahertz photons for several types of numerically integrated interaction were reported to emphasize the importance of the cascaded configuration concept. Cascaded OR processes in nonlinear optical crystals for intense terahertz pulse generation was also reported [15]. The authors considered both the perfect phase matching without loss and the realistic phase matching with linear absorption in ZnTe crystals. Terahertz field enhancement by using OR process with multiple pumping pulses was also studied. However, nonlinear absorption, which could severely limit the maximum THz conversion efficiency, was not taken into account [15]. For generating a tailored terahertz-frequency field, it is interesting to note that Koehl, *et al.* had

demonstrated the coherent superposition of the phonon-polariton wave packets in a ferroelectric crystal [16]. By using two temporally and spatially separated ultrafast optical excitation pulses, the researchers can control the propagating lattice excitations of a ferroelectric crystal and in principle tailor the terahertz-frequency fields that are radiated by the phonon-polariton wave packets.

Among many nonlinear optical crystals, GaSe is particularly suitable for terahertz generation owing to its high second-order nonlinearity and wide transparency range [17]. A narrow band DFG terahertz radiation source with a wide tuning range had been created by use of a 2-cm long GaSe crystal [8]. Femtosecond terahertz pulses tunable up to 41 THz had also been generated with a 90- μm thick phase-matched GaSe OR stage [6]. However, to further increase the output pulse energy of THz radiation by increasing the crystal length encounters a serious difficulty related to the pulse walk-off effect, which limits the effective interaction length in a nonlinear crystal. As a result, the useful length of GaSe for the generation of ultrashort terahertz radiation is typically in the order of millimeter. To overcome the difficulty, we reported in this study the successful generation of single-cycle high-amplitude terahertz radiation pulses with cascaded GaSe optical rectification stages.

2. Theoretical model and experimental methods

The nonlinear interacting processes between the optical and terahertz pulses in cascaded GaSe OR stages can be adequately described with the coupled wave equations. Under the slowly-varying envelope approximation, the coupled wave equations in frequency domain are given by [15]:

$$\frac{d}{dz} E_{T,2} = -j \frac{\epsilon_0 \mu_0 c d_{\text{eff}} \omega_{T,2}}{n(\omega_p)} \int E_p(z, \omega_p + \omega_{T,2}) E_p^*(z, \omega_p) \exp(j\Delta k_{T,2} z) d\omega - \frac{\alpha_{T,2}}{2} E_{T,2}(z, \omega_{T,2}), \quad (1)$$

$$\frac{d}{dz} E_p = -j \frac{\epsilon_0 \mu_0 c d_{\text{eff}} \omega_p}{n(\omega_{T,2})} \int E_p(z, \omega_p - \omega_{T,2}) E_{T,2}(z, \omega_{T,2}) \exp(j\Delta k_p z) d\omega - \frac{\alpha_p}{2} E_p(z, \omega_p), \quad (2)$$

where z is the propagation distance of optical and THz waves in GaSe; $E_{T,2}$ and E_p denote the terahertz radiation field and the optical pump wave in the second-stage GaSe crystal; ϵ_0 and μ_0 are the permittivity and permeability of free space, respectively; c is the speed of light in vacuum; d_{eff} is the effective nonlinear susceptibility of GaSe; $\omega_{T,2}$ and ω_p are the angular frequencies of the terahertz and the optical pump waves; $n(\omega_{T,2})$ and $n(\omega_p)$ are the refractive indices at the corresponding frequencies. $\alpha_{T,2}$ and α_p are the linear absorption coefficients of the terahertz and the optical waves of GaSe, respectively; $\Delta k_{p, T,2}$ denotes the wave-vector mismatch between the pump and terahertz waves. The total terahertz field in time domain can be described by:

$$E_T = E_{T,1} \exp(j\omega_{T,1} \tau) + E_{T,2}, \quad (3)$$

where $E_{T,1}$ and $E_{T,2}$ are the terahertz radiation fields from the first and the second OR stage, respectively; τ denotes the propagation delay time between the two OR stages.

The pure GaSe crystals used for this study were grown by the Bridgman method [18]. Raw materials were loaded in a well-cleaned quartz tube. The tube was then sealed and pumped down to below 10^{-6} Torr. The crystal growth was initiated with a thermal gradient of $30^\circ\text{C}/\text{cm}$ and a growth rate of 2 cm/day. The experimental setup for our multi-stage OR is shown in Fig. 1. The pump laser was a 1-kHz amplified Ti: Sapphire laser with pulse energy of 700 μJ and duration of 270 fs. The typical average pump power on the GaSe crystals was about 130 mW and 150 mW for the first and second stage, respectively. The pump beam diameter was adjusted to be about 3 mm for both stages. The GaSe crystals were configured for non-phase-matched OR [19]. The terahertz radiation field generated from the first OR stage with a 2-mm thick GaSe crystal was guided to the second OR stage with a 3-mm thick GaSe crystal. The terahertz pulses from the two OR stages were aligned collinearly with two gold-coated

parabolic mirrors. We blocked the residual 800 nm laser beams with teflon plates. An indium-tin-oxide (ITO) glass plate, which can transmit the 800-nm laser pulses while partially reflects the terahertz radiation, was used as the beam combiner. The time delay between the two terahertz pulses was carefully controlled with a translation stage. An optical chopper was used in this experimental arrangement to simultaneously modulate the optical pump beams for the first and the second GaSe OR stages. For monitoring the time-domain waveform of terahertz radiation, we employed the electro-optical sampling technique [20] with a 1-mm thick ZnTe crystal. Terahertz radiation generated from either the first or second stages or both can be recorded without moving any optical element.

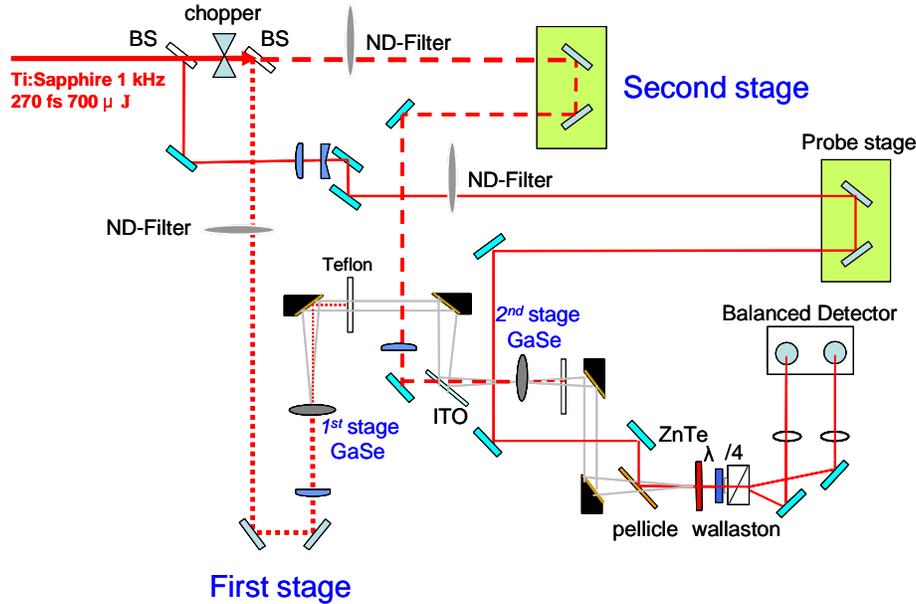


Fig. 1. Schematic of coherent generation of terahertz radiation by multi-stage optical rectification in GaSe crystals BS: Beam splitter; ND-Filter: Neutral Density filter; ITO: indium-tin-oxide glass plate; $\lambda/4$: quarter wave plate.

3. Results and discussions

We first adjusted the terahertz pulses from the two GaSe OR stages such that they overlapped spatially. The optical path length of the pump beam to the first GaSe stage was then varied to adjust the arrival time of the terahertz pulse at the second GaSe stage.

In Fig. 2, the curve of filled circles shows that the superposed waveform $E_T = E_{T,1} + E_{T,2}$ of terahertz fields from the cascaded GaSe OR stages with zero relative time delay. The curve of open squares was acquired with the same setup by blocking the pump pulse to the second GaSe crystal and scanning the delay time of the probe beam for the THz-TDS apparatus. The curve, which was scaled to allow for a direct visual comparison, can be used to properly reflect the injecting terahertz waveform from the first stage. The observed similarity of the two waveforms indicates a fairly high degree of mutual coherence existing between $E_{T,1}$ and $E_{T,2}$. Detailed information about the mutual coherence of $E_{T,1}$ and $E_{T,2}$ can be retrieved by repeating the measurement while varying the relative time delay τ .

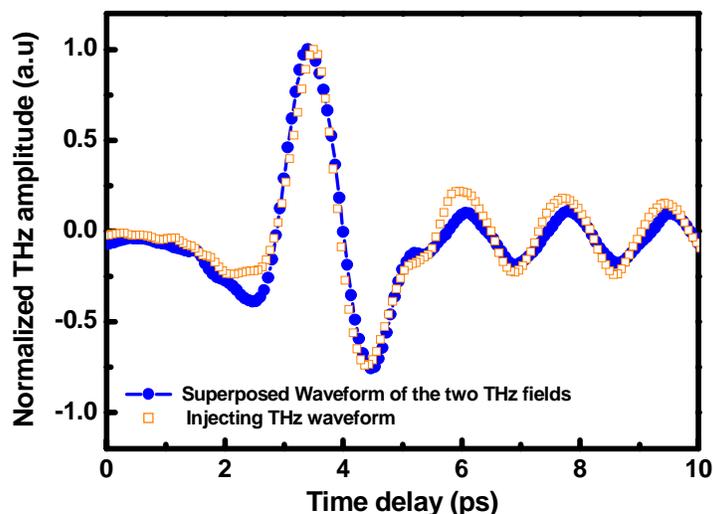


Fig. 2. Curve of filled circles shows the superposed waveform $E_t = E_{t,1} + E_{t,2}$ of terahertz fields from the cascaded GaSe OR stages with zero relative time delay. The curve of open squares represents the injecting terahertz waveform $E_{t,1}$ from the first stage and is properly scaled to offer a direct visual comparison.

By adjusting the time delay, the terahertz fields from the two OR stages can be superposed so that they interfere either constructively or destructively. To generate maximum terahertz field, the time delay of the two terahertz pulses should be adjusted for the best temporal overlap within 0.1 ps in our study, to yield constructive superposition over the entire spectral components involved. Figure 3 shows the terahertz time-domain waveforms and the spectra at three different time delays between the two OR stages. In these figures, the signals from the first and second stages by themselves are presented as black dashed and red dashed-dot lines, respectively. The superposed THz pulses from both stages are shown as the blue curve with open squares.

In Fig. 3(a), the main peak of terahertz field from the second OR stage leads that from the first stage. However, the trailing part of the THz pulse from the second stage still overlaps and interferes with the THz field from the first stage. The coherent superposition nature is more clearly revealed in the frequency domain shown in the inset of the Fig. 3(a). Parts of the terahertz spectral components from the two stages interfere constructively to produce higher spectral power while some spectral regions superpose destructively to yield lower spectral power. Thus the coherent superposition with multiple terahertz radiation sources offers a potential for the synthesis of terahertz field. In Fig. 3(c), the terahertz pulse from the second stage lags behind that from the first stage in such a way that the main positive peak from the second stage overlaps with the negative portion of the THz pulse from the first stage. The destructive superposition yields a spectrum shown in the inset of Fig. 3(c). The destructive superposition is caused by out-of-phase mixing of the terahertz field from the first stage with the optical pump pulse in the second OR stage. In other words, when pump pulse in the second OR stage and terahertz field from the first stage partly overlap in the time domain, the injecting terahertz field will dominate the three-wave mixing process and result in the output terahertz field profile variations. In the case of Fig. 3(c), the phase difference almost equals to π in the overlapped region between terahertz field from the first stage and the pump pulse in the second OR stage. Thus the terahertz field generated by the pump pulse in the second OR stage is then superposed with the first THz field to yield a much weaker terahertz radiation output.

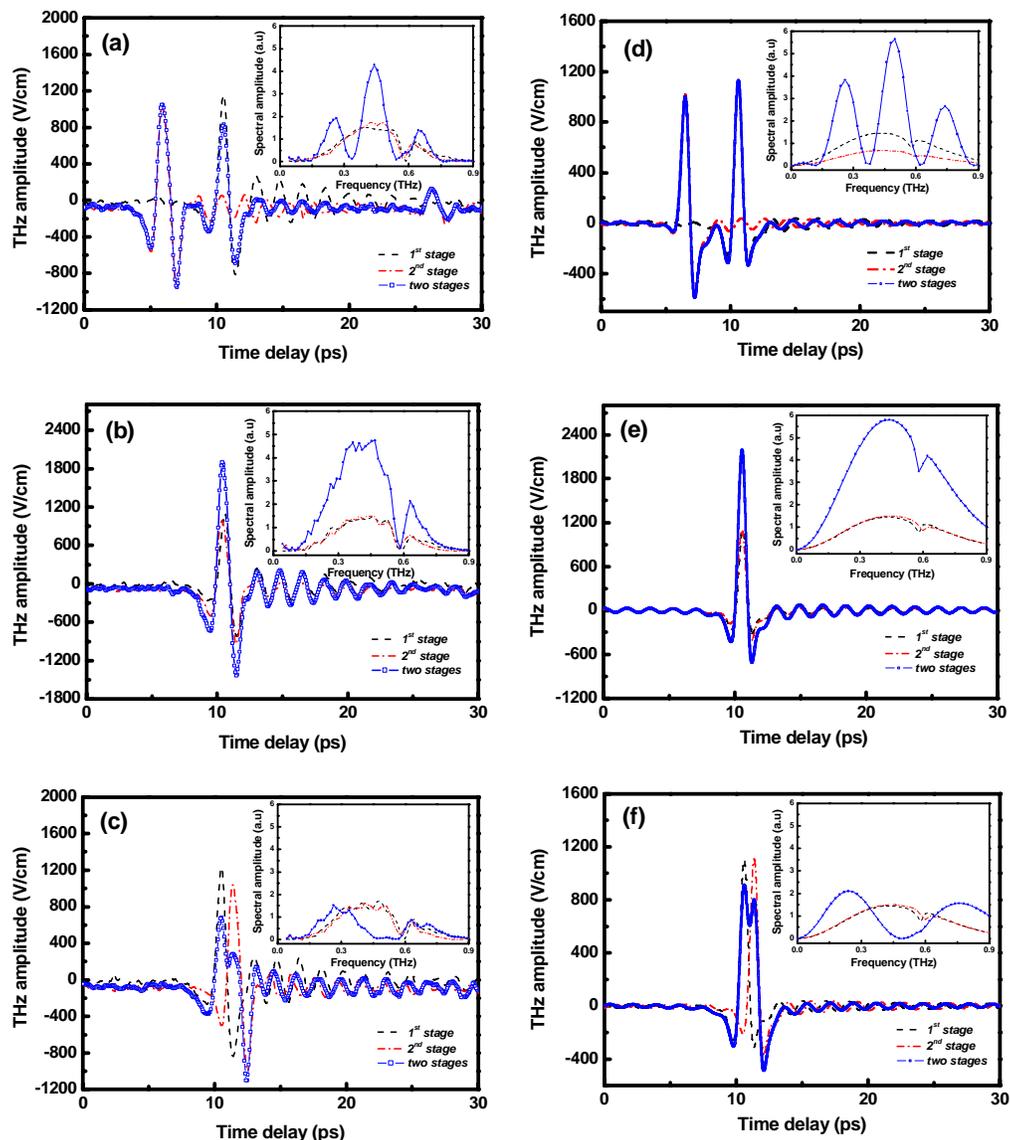


Fig. 3. Terahertz time-domain waveforms and spectra for three different time delays between the pump pulses of the first and second stages. (a) (b) (c) represent the experimental data, and (d) (e) (f) show the theoretical simulation results. In (a) and (d), the terahertz pulse from the second stage leads the signal from the first stage; in (b) and (e), the terahertz pulses from the first and second stages are overlapped in the time delay; in (c) and (f), the terahertz pulses from the second stage falls behind that from the first stage. Inset: corresponding spectra of the terahertz radiation.

The highest terahertz field amplitude can be obtained by synchronizing the first and second OR stages to attain constructive superposition of terahertz fields in the second OR stage. This can be done by injecting terahertz field with the correct phase at a proper arrival time relative to the optical pump pulse of the second stage. The output terahertz field

possesses the property of the injecting terahertz field but with higher amplitude. The inset of Fig. 3(b) presents the corresponding spectra of the terahertz radiation fields.

We have performed a theoretical simulation of the output of the two-stage OR with Eqs. (1)-(3). To allow for a straightforward comparison of the THz spectral profile with the measured data, an optical pump pulse with a spectral width < 1 THz is assumed in the numerical simulation. The absorption by the optical phonon mode of GaSe at 0.58 THz was also included in our simulation. The calculated results corresponding to the three different experimental conditions are presented in Figs. 3(d), 3(e), and 3(f) agree well with the experimental data shown in Figs. 3(a), 3(b) and 3(c). This confirms that the theoretical model used to depict the coherent multi-stage optical rectification processes in GaSe crystals is satisfactorily accurate.

The three-wave mixing phenomena can be appropriately explained with the theoretical model no matter what the injecting terahertz field is weak or strong. In fact, we have performed the theoretical calculation to verify the modulation of optical pump pulse by a weak injecting terahertz field. We found that the optical pump pulse can readily be modulated by the injecting terahertz field as long as the injection intensity is above the level of 3 orders of magnitude weaker than that of optical pump pulse. As that happens, the central wavelength of the output pump pulse red-shifts from that of the input pump pulse.

It is well known that linear and nonlinear optical absorption in GaSe have significant influences on the THz generation process [21-23]. Free carriers can be generated via two-photon absorption of GaSe pumped by high-intensity near infrared pulses. The terahertz radiation pulse propagating in GaSe can then be attenuated by free carriers. To access the free carrier absorption effect at THz frequencies, we placed the optical chopper in our setup (see Fig. 1) at a different position such that only the probe and the first pump beam were modulated by the chopper. In this way, the THz radiation generated by the second GaSe crystal will not be detected. We measured the peak amplitude of the terahertz radiation field while varied the optical pump intensity of the second GaSe OR stage. The results, as shown in Fig. 4, reveal that the measured terahertz radiation field decreases when the optical pump intensity is higher than 2 GW/cm^2 . The decreasing THz radiation must be caused by free carrier absorption occurring in the second GaSe crystal.

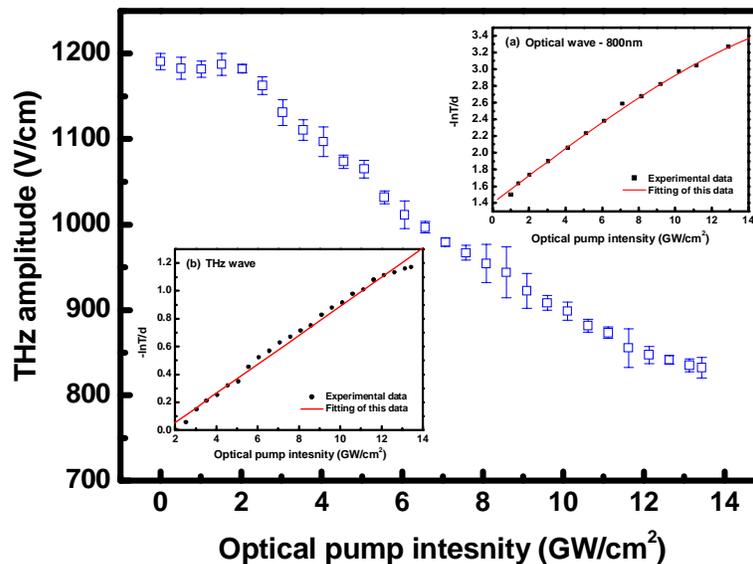


Fig. 4. Attenuation of THz radiation by the GaSe crystal under high intensity pump laser pulses. Inset (a): fitting of the experimental data for linear and nonlinear absorption coefficient of GaSe crystal pumped by 800 nm optical pulses. Inset (b): fitting of the experimental data for the absorption coefficient $\alpha_{\text{THz,fc}}$ at terahertz frequency in GaSe crystal due to free carriers.

The influence of multi-photon absorption on the generation of terahertz pulses by optical rectification had been revealed recently [24]. From our measurements, the linear and the nonlinear two-photon, three-photon absorption coefficients of optical wave at 800 nm in GaSe were determined to be $\alpha=1.40 \text{ cm}^{-1}$, $\beta=1.63 \times 10^{-10} \text{ cm/W}$, and $\gamma=3.99 \times 10^{-22} \text{ cm}^3/\text{W}^2$, respectively (see the inset (a) of Fig. 4). These are comparable to the values published in previous works [21, 22]. We can further estimate the number density of free carriers N in GaSe using [23]:

$$N = \left[\frac{F'}{\hbar\omega} \right] \left(\alpha + \beta \frac{F'}{\tau_p 2\sqrt{2}\sqrt{\pi}} + \gamma \frac{F'^2}{8\pi\tau_p^2} \right), \quad (4)$$

where $F'=F_0(1 - R)$ with F_0 denoting the energy density of the optical excitation; $\hbar\omega = 1.55 \text{ eV}$ ($\lambda=800 \text{ nm}$); the optical reflectivity at 800 nm $R=0.23$; and $\tau_p = \tau_{FWHM} / \sqrt{2 \ln 2}$ for the pump pulse with pulse width τ_{FWHM} . Under our experimental conditions, the number density of electron-hole pairs was estimated to be $N = 9.2 \times 10^{15} \text{ cm}^{-3}$ with $F=7 \text{ GW/cm}^2$.

The attenuation of terahertz radiation by the free carriers generated by two-photon absorption in GaSe at high optical excitation level can be calculated with

$$T = \frac{I}{I_0} = \exp(-\alpha_{THz,fc} d) . \quad (5)$$

Here I_0 and I denote the terahertz intensity before and after the GaSe crystal under test; d is the crystal thickness; and $\alpha_{THz,fc}$ is the absorption coefficient at terahertz frequency, defined as

$$\alpha_{THz,fc} = \sigma_{THz} N \quad (6)$$

with N being the free-carrier concentration and σ_{THz} the absorption cross-section of GaSe at terahertz frequency in the presence of free carriers. We can determine the absorption cross-section σ_{THz} by fitting the measured data to Eqs. (4) - (6). The results are presented in the inset (b) of Fig. 4. The deduced value of σ_{THz} lies in the range of $(5.4 - 6.3) \times 10^{-17} \text{ cm}^2$. Note that Kulibekov, *et al.* [23] had reported the absorption cross-section by free carriers at the optical wavelength $\lambda=626 \text{ nm}$ to be about $(4 - 6) \times 10^{-19} \text{ cm}^2$ [23]. Compared to the optical pulses, the spectra of terahertz pulses center at much lower photon energies ($E = 4 \text{ meV}$ for 1 THz), which are in the range of the binding energy of the excitons in many semiconductors. As a result, the weak terahertz signals are typically absorbed strongly by the photo-excited free carriers. Accordingly, it is reasonable that the value of absorption cross-section for the free carriers at the terahertz frequencies measured in this work is two orders of magnitude larger than that published in Ref. [23].

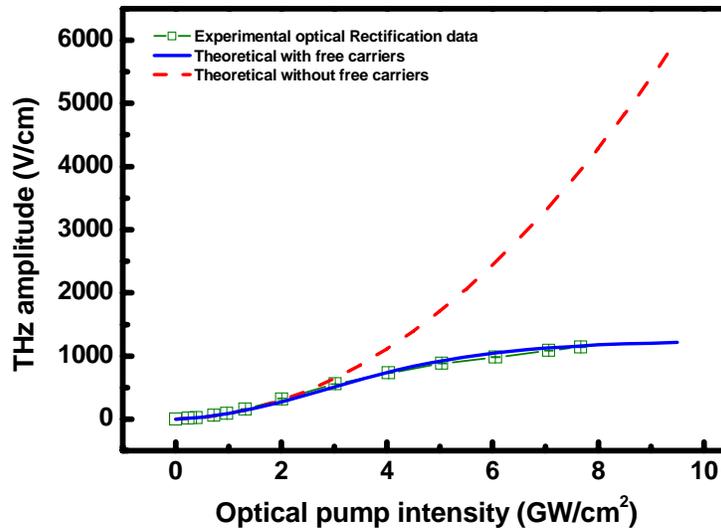


Fig. 5. Peak field amplitudes of the terahertz pulse at the output of the second OR stage are plotted as a function of pump intensity. Red and blue curves are theoretical predictions without and with the free-carrier absorption taken into account.

We further examined the power dependence of the terahertz radiation generated by optical rectification. Figure 5 shows the terahertz radiation output from the second OR stage without injecting as a function of optical pump intensity. A sub-linear dependence was found when the pump intensity is above 2 GW/cm². This can be attributed to the absorption of terahertz radiation by the free carriers generated by the pump pulses at 800 nm. All the calculation results were re-scaled properly in order to compare with the experimental data. We modeled the OR-generated terahertz radiation with Eq. (1). The red curve in Fig. 5 indicates the terahertz output by taking into account the linear optical absorption effect only. The terahertz output increases significantly with the pump intensity. When free carriers generated from the linear, nonlinear two-photon and three photon absorption were taken into account in the calculation, the result is presented as the blue-colored curve in Fig. 5. Clearly, the terahertz output saturates at high pump level, agreeing well with our measured data. The excellent agreement also indicates that the free carrier absorption effect places an upper limit on the terahertz radiation intensity at high pump levels.

Saturation of the terahertz radiation generated by OR from ZnTe crystal at high pumping level had also been reported and was attributed to two-photon absorption of the optical excitation beam [25, 26]. The limitation on terahertz radiation output by two-photon absorption can be alleviated to some extent by using larger beam size. The generation of high power terahertz radiation is then limited mainly by the finite interaction length of nonlinear optical process used.

The GaSe crystal thicknesses used in this work are 2 mm and 3 mm for the first and second OR stages, respectively. The pumping level for both stages is about 7 GW/cm². By using instead one OR stage with a single 5-mm-thick GeSe crystal pumped at 14 GW/cm², the longer interaction length simply produces more free carriers and causes more power loss of terahertz radiation output. We estimated the peak electric field of the terahertz radiation output from the single OR stage with a single 5-mm-thick GaSe crystal to be 1224 V/cm. This value is only one half of that from two coherently coupled OR stages with the total crystal lengths of 5 mm, which yields an output of 2285 V/cm without optimization.

Intense table-top terahertz radiation source had been demonstrated with large-aperture photoconductive (PC) antennas [27]. On the other hand, the pulsed high voltage need for biasing the PC antenna produces excess electrical noise. Further, the terahertz output saturates

at relatively low excitation fluence. Terahertz pulses with high peak electric fields can also be produced by four-wave mixing in air plasma [9]. In this method, high pump intensity up to TW/cm^2 is needed to effectively generate high-order nonlinear process in the plasma. Coherently coupled OR stages reported here offers an alternative for high-intensity THz pulse generation at lower pump intensity than that used in plasma THz source. In comparison to large-aperture photoconductive switches, the OR THz generation scheme produces broad-bandwidth THz pulses, which may be more suited for time-resolved spectroscopic applications. Micro-joule level terahertz pulse had been reported by using a large OR emitter [28]. Micro-joule level terahertz pulse could be actually obtained by enlarge the pump pulse diameter and upscale the pump pulse energy [5, 28]. In practice, the conversion efficiency of terahertz generation is usually limited by Manley-Rowe relation to the order about 10^{-6} - 10^{-4} [5, 8, 9, 13, 27, 28]. Conversion efficiencies much higher than the Manley-Rowe limit had been theoretical verified by cascaded difference frequency generation processes [14, 15]. Therefore, high conversion efficiency can be achieved with multi-stage OR scheme. The multi-stage OR technique can overcome in principle the limitations on the finite interaction length and generate higher terahertz field amplitude than that from one thick crystal. The price paid for the multi-stage generation of terahertz radiation is a more complex set-up and precise alignment between stages.

4. Conclusions

Generation of single-cycle terahertz pulses by two-stage optical rectification using GaSe crystals was proposed and experimentally demonstrated. By properly adjusting the time delay between the pump pulses to the two OR stages, the terahertz radiation field generated by the second stage can be constructively superposed to the injecting terahertz field from the first stage. The high mutual coherence between the two terahertz radiation fields is ensured with the coherent optical rectification process and can be further used to synthesize a desired spectral profile of output coherent THz radiation. Specifically, it is observed that free carrier absorption effect places an upper limit on the terahertz radiation intensity at high pump levels. Free carriers induced nonlinear absorption of THz radiation is further investigated in this study. The absorption cross-section, σ_{THz} , of GaSe crystal at terahertz frequency in the presence of free carriers is estimated in the range of $(5.4 - 6.3) \times 10^{-17} \text{ cm}^2$. This multi-stage OR technique can be useful for the generation of single-cycle high-amplitude terahertz pulse, which is not limited by the pulse walk-off effect from group velocity mismatch in the nonlinear optical crystal used.

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