Generation of broadband THz pulses in organic crystal OH1 at room temperature and 10 K

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Abstract: We studied the effects of cryogenic cooling of a 2-[3-(4-hydroxystyryl)-5, 5-dimethylcyclohex-2-enylidene] malonitrile (OH1) crystal on the generation of broadband THz pulses via collinear optical rectification of 1350 nm femtosecond laser pulses. Cooling of the OH1 crystal from room temperature to 10 K leads to a ~10% increase of the pump-to-THz energy conversion efficiency and a shift of the THz pulse spectra to a higher frequency range. Both effects are due to the temperature variation of the THz absorption and the refractive index of the OH1 crystal. This conclusion has been verified by temperature dependent measurements of the linear absorption in the THz frequency region. An approach to obtain a stronger increase of the THz generation efficiency at cryogenic cooling of the OH1 crystal is discussed.

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1. Introduction

Generation of high energy ultrashort THz pulses [1] remains an active area of research due to the current and potential applications in nonlinear spectroscopy [2, 3] including ultrafast magnetization dynamics study [4], characterization of X-ray and electron ultrashort pulses [1, 5–8], and biomedical imaging [9]. Today, high energy near single-cycle THz pulses can be obtained at large scale accelerator facilities (up to 600 μJ [1]) or by optical rectification of femtosecond laser pulses with tilted pulse fronts (TPF) in lithium niobate (up to 125 μJ [10]) [11, 12]. The latter method represents a tabletop technique. In addition, the generation of ≥460 μJ single cycle pulses with an average frequency of 0.4 THz from solid density plasmas created by a tabletop multi-terawatt femtosecond laser system was recently reported [13]. The average frequency of THz pulses delivered by TPF sources is typically below 1.5 THz. The strong increase in absorption in LiNbO3 above 1.5 THz prohibits efficient THz generation at these frequencies. Cryogenic cooling of LiNbO3 leads to a significant rise (by a factor of about 3.5) of the THz generation efficiency [12, 14, 15]. However, cooling does not allow substantially increasing the average frequency of generated THz pulses. To cover the frequency range between 1 and 10 THz, optical rectification in organic salt crystals (DAST, DSTMS, OH1 etc.) have been proven to allow for a high pump-to-THz conversion efficiency [14–17]. Recently, the generation of 45 μJ near single cycle pulses with an average frequency of 2.1 THz and 15 μJ pulses with an average frequency of 2.65 THz has been demonstrated by this technique [16, 19]. It was found that in order to obtain a high pump-to-THz energy conversion efficiency (up to 2% [16]) in many organic salt crystals, the application of femtosecond laser pulses at 1.2–1.5 μm is required. Alternatively the use of a DAST/SiO2
multilayer structure for efficient generation of near single cycle pulses with an average frequency of 6 THz via collinear optical rectification of 800 nm femtosecond laser pulses was proposed [20].

Previously it was found that cryogenic cooling of the widely used organic salt crystal DAST does not result in strong changes of its absorption and refractive indices [21] in the THz frequency range. Consequently, cooling does not increase THz generation efficiency or modify the THz pulse spectra noticeably [22]. However, the effect of cryogenic cooling on other organic salt crystals like DSTMS and OH1, that are used for high power THz generation [16–19], has not been studied before. In this paper, we report the cryogenic cooling effect on THz generation in the 2-[3-(4-hydroxystyryl)-5, 5-dimethylcyclohex-2-enylidene] malononitrile (OH1) crystal via collinear optical rectification of 1350 nm femtosecond laser pulses.

2. Experimental set-up

The experimental set-up used for studying the effect of cryogenic cooling on THz generation is shown in Fig. 1. A 1 mm thin OH1 crystal with a 3 mm clear aperture was mounted on the cold finger of a closed-cycle helium cryostat equipped with polyethylene cyclic olefin copolymer (Topas) windows [23] for THz beam output. The crystal was irradiated by 1350 nm, 60 fs pulses coming from a high power OPA pumped by a TW power Ti:sapphire laser system [24]. The laser beam polarization was parallel to the crystallographic c-axis, which is the direction with the highest nonlinear susceptibility. In order to obtain a top-hat homogeneous pump intensity distribution on the crystal surface, a broad NIR beam coming from an OPA was passed through a 3 mm aperture placed before the cryostat. The spectra of generated THz pulses were measured by a commercial Michelson interferometer with a Golay cell as a THz power detector.

Fig. 1. Experimental setup.

Linear absorption spectra of an OH1 crystal over a broad THz frequency range were measured at temperatures between 5 K and 300 K using a commercial FT spectrometer.

3. Experiment results and discussion

Figure 2 depicts the THz transmission spectra of an OH1 crystal with the optical axis oriented parallel to the THz wave polarization. The open cycle lines in Fig. 2 present experimental data obtained at different temperatures between 5 and 300 K, whereas the solid line are experiment data fits. The transmission spectra were fitted with a Drude-Lorentz dielectric function using a RefFIT program [25]:

\[
\varepsilon(\omega) = \varepsilon_\infty + \sum_i \frac{\omega_p^2}{\omega_0^2 - \omega^2 - iY_0\omega},
\]

where \(\varepsilon_\infty\) is the high-frequency dielectric constant, which takes into account the contribution of all oscillators at very high frequencies and \(\omega_p^2, \omega_0^2, Y_0\) are respectively the plasma frequency, the transverse frequency and the linewidth (scattering rate) of the i-th Lorentz oscillator. The Drude term, which describes the response of the free charge carriers is the case. The complex transmission coefficient \(t\) is given by:
\( t(d) = \exp(\frac{\omega}{c} \sqrt{\varepsilon(\omega) \cdot d}) \), where \( d \) is the thickness of the sample. Finally, the (measured) transmission \( T \) reads: \( T = |\mathbf{p}|^2 \). The fit parameters are shown in Table 1.

**Fig. 2.** THz transmission spectra of an OH1 crystal with optical axis oriented parallel to the THz wave polarization.

**Table 1.** Fit parameters of the OH1 crystal transmission spectra [Fig. 2] to a Drude-Lorentz model.

| Temperature (K) | Oscillator frequency (THz) | Oscillator strength | Line Width (THz) | Temperature (K) | Oscillator frequency (THz) | Oscillator strength | Line Width (THz) |
|----------------|---------------------------|--------------------|-----------------|----------------|---------------------------|--------------------|-----------------|
| 300            | 1.45                       | 45.7               | 0.13            | 150            | 1.48                       | 39.7               | 0.08            |
|                | 1.58                       | 17.3               | 0.14            |                | 1.66                       | 16.2               | 0.12            |
|                | 2.23                       | 78.8               | 0.21            |                | 1.83                       | 32.8               | 0.74            |
|                | 2.41                       | 92.0               | 0.04            |                | 1.97                       | 3.8                | 0.07            |
|                | 2.71                       | 150.5              | 1.80            |                | 2.25                       | 45.7               | 0.13            |
|                |                            |                    |                 |                | 2.40                       | 53.7               | 0.05            |
|                |                            |                    |                 |                | 2.58                       | 96.8               | 0.47            |
|                |                            |                    |                 |                | 2.79                       | 87.85              | 0.28            |
| 250            | 1.47                       | 41.7               | 0.11            | 100            | 1.49                       | 35.9               | 0.07            |
|                | 1.59                       | 32.0               | 0.27            |                | 1.68                       | 11.6               | 0.10            |
|                | 1.84                       | 5.0                | 0.10            |                | 1.74                       | 27.7               | 0.51            |
|                | 1.94                       | 7.9                | 0.15            |                | 1.95                       | 7.5                | 0.15            |
|                | 2.24                       | 63.8               | 0.19            |                | 2.38                       | 81.3               | 0.24            |
|                | 2.41                       | 56.0               | 0.09            |                | 2.61                       | 54.5               | 0.19            |
|                | 2.60                       | 109.7              | 0.35            |                | 2.77                       | 38.0               | 0.12            |
|                | 2.85                       | 119.0              | 1.6             |                | 2.86                       | 36.8               | 0.09            |
| 200            | 1.47                       | 42.3               | 0.09            | 5              | 1.5                        | 22.1               | 0.06            |
|                | 1.63                       | 21.0               | 0.18            |                | 1.74                       | 9.8                | 0.09            |
|                | 1.82                       | 8.9                | 0.18            |                | 1.83                       | 30.8               | 0.66            |
|                | 1.97                       | 10.1               | 0.23            |                | 2.05                       | 7.6                | 0.12            |
|                | 2.27                       | 62.5               | 0.16            |                | 2.38                       | 42.2               | 0.07            |
|                | 2.39                       | 58.5               | 0.12            |                | 2.53                       | 32.2               | 0.12            |
|                | 2.66                       | 105.4              | 0.36            |                | 2.67                       | 36.7               | 0.08            |
|                | 2.85                       | 107.4              | 1.27            |                | 2.87                       | 27.0               | 0.15            |
|                |                            |                    |                 |                | 2.96                       | 22.3               | 0.08            |
Figures 3(a) and 3(c) show typical interferograms of the THz pulses generated at room temperature and at 10 K via optical rectification of laser pump pulses with a moderate pulse energy (0.14 and 0.15 mJ). Corresponding FFT power spectra are shown in Figs. 3(b) and 3(d).

The spectra of THz pulses generated at different pump intensities at room temperature and at 10 K are presented in Figs. 4(b)–4(d). The overall THz pulse spectra are in good agreement with the THz linear absorption spectra of the OH1 crystal shown in Fig. 4(a) and with previously reported spectra of THz pulses generated in this crystal [26]. Unfortunately, the used Fourier spectrometer does not allow to measure THz transmission spectra [Figs. 2, 4(a)] of the OH1 crystal below 1.2 THz because of the small clear aperture (3 mm) of the available crystal. On the other hand, the interferometric measurements [Figs. 3(a), 3(c)] of spectra of the THz pulses generated in the OH1 crystal provide reliable information down to 0.17 THz [Figs. 3(b), 3(d), 4(b)–4(c)], due to a sampling window of 6 ps. The dip at about 1.5 THz in the pulse spectra corresponds to an absorption line at this frequency. Cryogenic cooling of the OH1 crystal leads to a power decrease and narrowing of this absorption line. Moreover, cooling results in an increase in the central line frequency. Such a thermal frequency shift was observed in a DAST crystal for the absorption line at 1.2 THz [21]. However, the absorption line at the central frequency of 3.4 THz in the DAST crystal shows neither a thermal frequency shift nor a narrowing with cryogenic cooling [22]. Despite a decrease in THz absorption in the OH1 crystal with temperature between 1.5 and 1.7 THz, we did not observe an increase in THz pulse spectral power in this frequency range. This phenomenon probably originates from the phase mismatch appearing at these frequencies upon cooling, as the phase refractive index at these frequencies decreases relative to the group refractive index at 1350 nm. The same effect leads to a decrease in THz pulse power in the frequency range of 0.5–1.3 THz. The decrease in THz pulse spectral power with frequency rise above 2 THz at room temperature results from a rise in absorption and phase mismatch related to a strong and broad absorption band centered at about 3 THz. An increase in THz pulse power in the 2–2.2 THz spectral range observed upon cooling is determined by two effects: a decrease in THz absorption and better phase-matching at these frequencies due to a narrowing of the absorption band at 3 THz. In previous experiments at room temperature [26], a small increase in THz spectral power in this frequency range could be achieved by improved phase matching by tuning the pump laser wavelength from 1400 nm to 1250 nm.

In addition, the low frequency part of the THz pulse spectra (centered at 1.25 THz at room temperature) rises faster than the high frequency part (centered at 1.8 THz at room temperature) with pump intensity.
In particular, this effect is noticeable at room temperature [Figs. 4(b)-4(d)]. This effect can appear due to a spectral narrowing of the laser pump pulses due to efficient THz generation [27]. The spectral narrowing leads to a decrease of the THz generation efficiency at the high frequency part of the spectra. From the spectral measurements (Figs. 4(b)-4(d)) we have estimated that by cooling the OH1 crystal from room temperature to 10 K the THz pulse energy is increased by 10%. This increase is a relatively small improvement as compared to the THz generation efficiency rise observed in lithium niobate at cryogenic temperatures. The difference is probably related to the weaker phonon-phonon coupling in organic crystals, which leads to a relatively weak THz absorption and thus has a relatively limited potential to change in absolute value with temperature. More significant increase of the THz generation efficiency in OH1 crystal can probably be achieved by cryogenic cooling combined with tuning of the central wavelength of femtosecond laser pump pulses. However, proper tuning of the laser wavelength requires an additional study of the temperature dependencies of THz and near infrared refractive indexes of the OH1 crystal.

4. Conclusion

We have experimentally shown that the generation efficiency of ultrashort THz pulses via collinear optical rectification of 1350 nm femtosecond laser pulses in OH1 can be increased by 10% through cryogenic cooling. Moreover, a shift in the THz pulse spectrum towards the higher frequency range at the crystal cooling was observed. Whereas the THz absorption in OH1 crystal is noticeably reduced by cryogenic cooling, phase mismatching appearing due temperature variation of the THz refractive index is limiting the conversion efficiency. A stronger increase of the THz generation efficiency can possibly be achieved by cryogenic cooling of the OH1 crystal complemented by proper tuning of the laser pulse wavelength.

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