Theoretical Study of Organic Crystal-Based Terahertz-Wave Difference Frequency Generation and Up-Conversion Detection

Pengxiang Liu $^{1,2,3}$ · Feng Qi $^{1,2,3}$ · Weifan Li $^{1,2,3}$ · Zhaoyang Liu $^{1,2,3}$ · Yelong Wang $^{1,2,3}$ · Wei Shi $^4$ · Jianquan Yao $^4$

Received: 20 April 2018 / Accepted: 20 June 2018 / Published online: 3 July 2018
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Abstract Optical pumped quasi-monochromatic and tunable terahertz (THz) wave generation and detection system is investigated theoretically. Calculations are performed on an organic nonlinear medium: 4'-dimethylamino-N-methyl-4-stilbazolium tosylate (DAST), due to the superior properties (large dynamic range and extremely wide tunability). The behavior of pulses during the nonlinear interaction is characterized. Previous experimental results are well explained. Our theoretical model provides an approach to improve the performance of the coherent THz system from two aspects. For the source part, an optimal crystal thickness that maximizes the THz output is obtained. For the detector part, a linear conversion relation (used for calibration) and the parameter dependence of responsivity are provided. It was predicted that a dynamic range of ten orders could be achieved with the DAST-based THz system. The presented model can also be applied to guide the design of THz systems with other nonlinear mediums in collinear phase-matching geometry.

Keywords Nonlinear optics · Parametric process · Terahertz-wave generation · Terahertz-wave detection · Organic nonlinear crystal

Feng Qi
qifeng@sia.cn

1 Shenyang Institute of Automation, Chinese Academy of Sciences, Shenyang 110016, China
2 Key Laboratory of Opto-Electronic Information Processing (CAS), Shenyang 110016, China
3 Key Lab of Image Understanding and Computer Vision (Liaoning Province), Shenyang 110016, China
4 College of Precision Instruments and Opto-Electronics Engineering, Tianjin University, Tianjin 300072, China
1 Introduction

Terahertz (THz) spectroscopy and imaging technology with high resolution have attracted considerable interest in a variety of applications [1], such as bio-medical testing, nondestructive evaluation, and environmental monitoring. Widely tunable and monochromatic THz sources play a key role in these fields. Optical pumped coherent THz sources based on nonlinear frequency down-conversion (difference frequency generation (DFG) [2, 3] and stimulated polariton scattering (SPS) [4]) are well suited for the application systems, due to the laser-like linewidth and beam quality [5], continuous and wide tunability [6], and operating at room temperature. However, the optical THz sources commonly operate in ns-pulses with low repetition rate (extremely low duty cycle) [2, 4, 6]. As a result, the low average output power as well as the poor sensitivity of traditional thermal detectors greatly limits the dynamic range of THz analysis system. Besides, the THz pulse envelope (in the order of ns) cannot be captured by these detectors with small bandwidth (~ 100 Hz of Golay cell and ~ 30 kHz of bolometer).

Coherent THz detection can be achieved with electro-optic sampling in the time-domain spectroscopy (TDS) [7]. Similarly, up-conversion detection for the ns THz pulses based on nonlinear optics was also developed [8]. High sensitivity and fast response time at room temperature can be provided, which benefit from the advanced detection technologies in optical region. This approach could well overcome the drawbacks of the optical THz sources, greatly increase the dynamic range of the measurement system, and contribute to real-time THz imaging [9, 10]. It was first demonstrated with GaP [11, 12] and later extended to other crystals, including GaSe [13], ZnGeP2 [14], periodically inverted GaAs [15], and LiNbO3 [16].

In the past decades, considerable works on organic crystal-based THz-wave generation and detection have been performed, because of the superior optical properties: high second-order nonlinearity and favorable type-0 phase-matching. Output of the sources covers a wide tuning range (up to 20 THz), e.g., with organic salt 4′-dimethylamino-N-methyl-4-stilbazolium tosylate (DAST) [6]. On the other hand, low noise equivalent power (NEP) [17] and wide detectable frequency band [18] were achieved. Thus, organic crystal is proved to be a promising medium for THz-wave nonlinear frequency mixing and potential to be applied in the systems. However, the theoretical description of this physical process has still not been studied sufficiently. As a kind of indirect detection, the conversion relations between the parameters (including powers and pulse widths) of incident THz wave and up-converted near-infrared (IR) signal are necessary for the calibration. Recently, theoretical models of noncollinear SPS in LiNbO3 (steady-state [19] and time-dependent [20]) were reported.

In this paper, we present a comprehensive study of pulsed THz-wave difference frequency mixing in collinear geometry. The theoretical model is based on time-dependent coupled wave equations, which takes into account the pulse envelopes of three interactive waves. The dynamic of optical-to-THz energy conversion is investigated, to find an optimal generation length. In the part of detection, we go further and derive an analytical relation between IR output and THz input, which is compared with numerical results. The response of DAST-based detector to different THz frequencies is discussed.

2 Theoretical Framework

Organic crystal-based THz-wave generation and detection system was illustrated in Fig. 1. Laser beams with two close frequencies (ω1 > ω2) incident on one crystal (source part).
Monochromatic THz wave at difference frequency (\(\omega_T = \omega_1 - \omega_2\)) was generated via second-order nonlinear effect. The THz wave combined with a laser beam (\(\omega_p\)) and interacted within the other crystal (detector part). A new laser frequency in near-infrared region (\(\omega_{IR} = \omega_p - \omega_T\)) was delivered. By measuring the up-converted IR signal, the characteristics of THz wave can be analyzed. The output power of source and NEP of detector determine the dynamic range of measurement system.

Both the generation and detection processes are collinear phase-matched difference frequency mixing. The behavior of three interactive waves should obey the coupled wave equations (plane wave and ns-pulse) [21]:

\[
\begin{align*}
\left(\frac{\partial}{\partial z} + \frac{n_T \omega}{c \partial t}\right) A_T(z, t) &= -\frac{\alpha_T}{2} A_T(z, t) + \frac{i \omega_T d_{\text{eff}}}{n_T c} A_1(z, t) A_2^*(z, t) \exp(i \Delta k z) \\
\left(\frac{\partial}{\partial z} + \frac{n_1 \omega}{c \partial t}\right) A_1(z, t) &= -\frac{\alpha_1}{2} A_1(z, t) + \frac{i \omega_1 d_{\text{eff}}}{n_1 c} A_2(z, t) A_T(z, t) \exp(-i \Delta k z) \\
\left(\frac{\partial}{\partial z} + \frac{n_2 \omega}{c \partial t}\right) A_2(z, t) &= -\frac{\alpha_2}{2} A_2(z, t) + \frac{i \omega_2 d_{\text{eff}}}{n_2 c} A_1(z, t) A_T^*(z, t) \exp(i \Delta k z)
\end{align*}
\]

where \(A(z,t)\) is the time-dependent amplitude envelope, \(d_{\text{eff}}\) is the effective nonlinear coefficient of crystal (i.e., \(d_{11}\) of DAST = 210 pm/V [22]), and \(\Delta k = k_1 - k_2 - k_T\) is the wavevector mismatch. For the detection process, the subscripts 1 and 2 should be replaced by p and IR, respectively.

We consider that three pulses have the same group velocities (\(u\)), according to the properties of organic crystals: (i) the dielectric constants in optical and THz regions are close; (ii) the high nonlinearity leads to a short effective interactive length (~ 1 mm). Then, a coordinate transformation with \(\zeta = z\) and \(\tau = t - z/u\) can be applied [23]. The set of partial differential Eq. (1) is converted into ordinary differential equations:

\[
\begin{align*}
\frac{\partial}{\partial \zeta} A_T(\zeta, \tau) &= -\frac{\alpha_T}{2} A_T(\zeta, \tau) + \frac{i \omega_T d_{\text{eff}}}{n_T c} A_1(\zeta, \tau) A_2^*(\zeta, \tau) \exp(i \Delta k \zeta) \\
\frac{\partial}{\partial \zeta} A_1(\zeta, \tau) &= -\frac{\alpha_1}{2} A_1(\zeta, \tau) + \frac{i \omega_1 d_{\text{eff}}}{n_1 c} A_2(\zeta, \tau) A_T(\zeta, \tau) \exp(-i \Delta k \zeta) \\
\frac{\partial}{\partial \zeta} A_2(\zeta, \tau) &= -\frac{\alpha_2}{2} A_2(\zeta, \tau) + \frac{i \omega_2 d_{\text{eff}}}{n_2 c} A_1(\zeta, \tau) A_T^*(\zeta, \tau) \exp(i \Delta k \zeta)
\end{align*}
\]

In the following sections, we will perform numerical calculations on the dynamic of THz-wave generation and detection in DAST crystals, based on (2) and the corresponding boundary conditions at \(\zeta = 0\).
3 THz-Wave Generation

To optimize the source part, we first analyze the process of DFG. The difference frequency is set to be $\omega_T/2\pi = 4$ THz, close to an emission peak of DAST [6]. The input optical intensities are $I_{1,2} = 100$ MW/cm$^2$, each with a Gaussian temporal envelope and a full width at half maximum (FWHM) of 10 ns. The beams are assumed to be flat-top with a radius of $r = 0.3$ mm. The shorter pump wavelength is $\lambda_1 = 1.253 \mu$m, phase-matched with 4 THz. The refractive indices and absorption coefficients are given by [24, 25].

The pulse envelopes of pump and THz waves at different interaction length $\zeta$ are presented in Fig. 2. Strictly speaking, a distance of 0.5 mm corresponds to a time delay of 3.6 ps. Here, we separate the curves with a step of 10 ns to show them clearly. Otherwise, the curves would be overlapped. The depletion of $\lambda_1$ and growth of $\lambda_2$ can be observed. A dip arises at the pulse center of $\lambda_1$ for further energy conversion. Similar result has been reported experimentally by [4]. As a second-order nonlinear effect, the frequency conversion process (both the depletion and growth) is faster at the pulse center, due to higher power. Consequently, more power of $\lambda_1$ is consumed at the center.

There is an optimal conversion length for THz output (seen in Fig. 3). As interpreted by the universal chart for THz DFG [26], the optimal length is determined by several factors including the effective nonlinear coefficient, pump peak powers, absorption loss, and phase-matching condition. The well-known “coherent length” is one of these factors: phase-matching. The difference between two lengths has been discussed in [27]. In our case, THz peak power reaches the maximum 355.67 W at $\zeta = 0.85$ mm (solid curve) and pulse energy 2.74 $\mu$J at 0.9 mm (dashed curve). The highest energy and photon conversion efficiencies from $\lambda_1$ to THz are $9.13 \times 10^{-4}$ and 5.467%, respectively. Both the nonlinear coefficient and absorption coefficient of DAST are much larger than those of inorganic crystals, which leads to a small optimal length (below 1 mm). In a recent experiment [28], a photon efficiency of 2.9% was obtained with another organic crystal OH1 (1.9-mm thick). The highest value of ~40% was reported by [29] with 2.6-mm-thick GaP under several MW pump power.

![Fig. 2 Evolution of pulse envelopes of pump and THz waves in the process of DFG within a DAST crystal](image-url)
As discussed above, the frequency conversion process is not synchronized at different relative time delays around the pulse center. When $\zeta$ exceeds the optimal length, the THz power at the center starts to decrease and that at the edges is still rising, so the pulse width becomes wider (squares in Fig. 3). In some experimental studies [2, 6], THz pulse width was estimated to calculate the peak power. By considering the temporal envelope of THz output as the product of those of input dual wavelengths, we get a formula: $1/\tau_1^2 = 1/\tau_1^2 + 1/\tau_2^2$ (dashed and dotted line in Fig. 3). The squares almost follow the line in the initial stage. At $\zeta < 1$ mm, the error between numerical result and analytical estimation is negligible.

4 THz-Wave Detection

The difference between the detection and generation processes is the boundary conditions. For the detection process, one of the input waves ($\omega_T$) is much weaker than the other ($\omega_p$). The conversion efficiency is much lower, compared with that of generation. We treat the amplitude of pump as a constant ($\partial A_p/\partial \zeta = 0$), since little pump energy is consumed (dotted curve in Fig. 4). Then, analytical solutions of (2) can be derived:

$$A_{IR}(\zeta, \tau) = \frac{i\omega_{IR}d_{eff}}{n_{IR}c}A_pA_T^*(0, \tau)\frac{\exp(\beta_1\zeta) - \exp(\beta_2\zeta)}{\beta_1 - \beta_2}$$ (3)

$$A_T(\zeta, \tau) = A_T(0, \tau) \left[ \frac{\beta_1\exp(\beta_1\zeta) - \beta_2\exp(\beta_2\zeta)}{\beta_1 - \beta_2} \right]^* \exp(i\Delta k\zeta)$$ (4)
Here, the optical absorption loss ($\alpha_{\text{IR}}$) is ignored. $\beta_1$ and $\beta_2$ are the eigenvalues:

$$
\beta_{1,2} = \frac{i\Delta k}{2} - \frac{\alpha_T}{4} \pm \sqrt{\left(\frac{i\Delta k}{2} - \frac{\alpha_T}{4}\right)^2 + \frac{\omega_{\text{IR}}\omega_{\text{T}}d_{\text{eff}}^2}{n_{\text{IR}}n_{\text{T}}e^2} |A_p|^2}
$$

(5)

The solutions of the coupled wave equations for detection process are shown in Fig. 4. Here, the parameters of pump wave $\lambda_p$ are the same as those of $\lambda_1$ in Section 3. The input THz pulse is that generated at $\zeta = 0.85 \text{ mm}$ in Fig. 3 (maximal peak power). In Fig. 4, the solid curves are the peak powers of up-converted IR and THz waves, solved numerically with (2), and the dashed curves are plotted with (3) and (4). The analytical approximation above is reasonable, if the crystal is thin ($\zeta < 1 \text{ mm}$). The IR power grows monotonically. As seen in the inset of Fig. 4, THz power decreases first, due to the high absorption of DAST ($\alpha_T = 74.79 \text{ cm}^{-1}$ at 4 THz), and increases later, because of the gain in the nonlinear frequency mixing. The discrepancy between the dashed and solid curves becomes obvious for further energy conversion. The solid curve is more precise, because the pump depletion is taken into account.

Equation (3) indicates a linear relationship between the up-converted IR signal and THz input:

$$
P_{\text{IR}}|_{\text{out}} = \frac{2\omega_{\text{IR}}d_{\text{eff}}^2}{\varepsilon_0 n_{\text{IR}}n_{\text{p}}n_{\text{T}}c^3} \frac{P_p}{\pi r^2} \frac{|\exp(\beta_1 L) - \exp(\beta_2 L)|^2}{\beta_1 - \beta_2} P_T|_{\text{in}}
$$

(6)

where $P$ is the peak power and $L$ is the crystal thickness. The factor before $P_T$ on the right side of (6) can be regarded as the responsivity of up-conversion detector $R_{uc}$. High nonlinearity of crystal, high power density of pump, and large crystal thickness contribute to large responsivity.

$$
R_{uc} = \frac{2\omega_{\text{IR}}d_{\text{eff}}^2}{\varepsilon_0 n_{\text{IR}}n_{\text{p}}n_{\text{T}}c^3} \frac{P_p}{\pi r^2} \frac{|\exp(\beta_1 L) - \exp(\beta_2 L)|^2}{\beta_1 - \beta_2}
$$

(7)

\begin{figure}[h]
  \centering
  \includegraphics[width=\textwidth]{fig4.png}
  \caption{Variations of peak powers of pump, IR, and THz waves in the process of detection within a DAST crystal. The solid curves present the numerical solutions of (2), and the dashed curves are plotted with (3) and (4).}
\end{figure}
The input-output characteristics of detection process under three conditions are presented in Fig. 5. The circles, squares, and triangles are calculated numerically with (2), which are close to the three lines based on (6). The linear relationship agrees well with previous experimental results [30]. To estimate the minimum detectable (MD) power, we assume the IR detector to be a simple PIN diode with a NEP of $\sim fW/Hz^{1/2}$ and a bandwidth of 400 MHz and take into account the optical loss in the system (including that caused by the low-pass filter). The MD power of up-converted IR signal is set as 100 nW in the calculation. Under a pump intensity of $I_p = 100$ MW/cm$^2$ and the crystal thickness of $L = 1$ mm, the MD THz peak power is 14.61 nW. Considering the highest output power of source part (355.67 W), a dynamic range of ten orders could be achieved with the DAST-based THz system.

The MD power of 14.61 nW corresponds to a NEP of 0.73 pW/Hz$^{1/2}$. For a He-cooled bolometer, the NEP is in the order of 0.1 pW/Hz$^{1/2}$. However, it is calibrated by CW waves rather than short pulses. These two values cannot be compared directly. The relationship between the NEP of photodiode and that of up-conversion THz detector is given by:

$$NEP_{\text{THz}} = \frac{NEP_{\text{PD}}}{R_{\text{uc}} \times \eta_{\text{filter}}}$$

(8)

where $\eta$ is the filter efficiency. In our calculation, the factor $R_{\text{uc}}$ is 6.84 and the photon efficiency from THz to IR is 11.44%. If the single photon detection is applied to the up-converted IR signal, the MD number of THz photons should be 9.

Another advantage of the up-conversion detection is the fast response time. THz temporal envelope has been determined indirectly based on those of pump light and IR signal ($1/\tau_T^2 = 1/\tau_{IR}^2 - 1/\tau_p^2$ [13, 17]). Our theoretical model could also describe this characteristic. Figure 6a shows the pulse envelopes of input pump light (dotted), THz wave (solid), and output IR signal (dashed curve) of a 1-mm-thick DAST crystal. The parameters used here are the same as those of Fig. 4. The pump and THz pulses are with FWHMs of 10 and 7.275 ns. The IR pulse

![Fig. 5 Relationships between the input THz powers and output IR powers at different conditions. The scatter points are calculated numerically, and the lines are plotted with (6)](image)
width decreases during the process of frequency mixing (squares in Fig. 6b, calculated with (2)). The reason is similar to that of Fig. 3. At detection lengths $\zeta = 0.5$ and $1$ mm, the IR pulse widths are 5.813 and 5.627 ns, respectively. Compared with the formula above (dashed and dotted line), the corresponding relative deviations are 1.19 and 4.35%.

The response of detector to different THz frequencies is also worth to be investigated. The dashed curve in Fig. 7 shows the MD THz power by a 1-mm-thick DAST under 100 MW/cm$^2$ pump intensity and phase-matched wavelength. The peaks coincide with the absorption peaks of DAST ($\downarrow$). Low absorption coefficient contributes to low MD power and high detectivity. In some experiments [17, 30], filters with fixed passband were used. We also calculate the MD powers under fixed IR wavelength ($\lambda_{\text{IR}} = 1.3 \mu$m). The phase mismatch becomes obvious at frequencies above 8.4 THz. As seen in Fig. 7, fJ-level MD energy of THz pulse can be achieved.
in a band below 13 THz. Calculation is not performed at higher frequencies, due to the lack of property parameters.

Monochromatic and tunable sources have been applied in active THz systems for the fields such as molecular spectroscopy [31, 32]. Compared with the general TDS systems, these DFG-based systems covered wider frequency band and provided higher resolution. To improve the signal-to-noise ratio, He-cooled bolometer was utilized as the detector. As reported by [17], the up-conversion detection is even more sensitive than the cryogenic detectors, which would contribute to the all-room-temperature operation of active THz system. Besides, the presented detection technique could reduce the requirement of pump source and potentially operate at relatively low peak power. It is possible for the system to implement the compact sources in high repetition rate, e.g., passively Q-switched [33] or intracavity schemes [34].

5 Conclusion

To conclude, we presented a theoretical study of coherent THz-wave generation and detection via nonlinear optical frequency conversion. Numerical calculations were performed on a system of DAST crystals pumped by ns-pulses, comprehensively. The dynamic of energy conversion and evolution of pulse envelopes were described. For the source part, an optimal crystal thickness was obtained. For the detector part, the conversion relation between the incident THz power and up-converted IR power was provided. High nonlinearity of crystal, high power density of pump, and large crystal thickness contribute to large responsivity and low NEP. Our calculations were in accordance with previous experimental results. It was predicted that a dynamic range of ten orders could be achieved with the DAST-based THz system. The presented model can also be applied to other nonlinear mediums in collinear geometry, which allows us to improve the performance of optical pumped quasi-monochromatic THz system in the application fields.

Funding Information

This work is supported by the National Natural Science Foundation of China (No. 61505089, 61605235, 61735010, and 61335013), the National Key Research and Development Program of China (No. 2016YFC0102900), and Hundred Talent Program of Chinese Academy of Sciences.

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