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Citation: Chin. Phys. B, 2025, 34 (9): 094211. DOI: 10.1088/1674-1056/addbcd

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Chin. Phys. B, 2025, 34 (7): 074205. DOI: 10.1088/1674-1056/adc7f7

Twin pulses of THz generation from a nonlinear crystalline quartz by femtosecond laser pulse

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(Received 15 April 2025; revised manuscript received 15 May 2025; accepted manuscript online 22 May 2025)

We observed sub-picosecond terahertz (THz) pulses generated from an electro-optic quartz plate using femtosecond optical pulses. The time-resolved THz radiation signal clearly indicates two separated THz pulses with opposite polarity. Based on our results, a model based on optical-to-THz conversion via optical rectification is proposed to describe the twin pulsed THz emission mechanism of the quartz plate. Firstly, the two separated THz pulses are assigned to the forced and free THz pulses, respectively. With an optical pulse serving as an external source in the crystalline quartz, the forced THz pulse is a solution to linear Maxwell's equations and propagates at the velocity of the pump laser pulse, while the free THz pulse propagates with the group velocity in the THz frequency range. Finally, as a non-centrosymmetry material, the THz amplitude exhibits perfect threefold symmetry with respect to the azimuthal angle and twofold symmetry with respect to the pump polarization angle.

Keywords: crystalline quartz, terahertz emission, optical rectification effect, forced electromagnetic pulses

PACS: 42.65.Ky, 42.72.Ai, 71.36.+c, 42.65.Re

DOI: 10.1088/1674-1056/addbcd

CSTR: 32038.14.CPB.addbcd

1. Introduction

Terahertz (THz) radiation, in the frequency range between microwave and infrared, is a significant research field due to its unique properties and diverse applications.^[1,2] Its unique properties, especially non-ionizing penetration through non-conductive materials such as polymers,^[3] ceramics,^[4] and biological tissues,^[5] make it ideal for non-invasive imaging and spectroscopic analysis.^[6,7] In addition, THz waves show great promise in advanced fields such as superconductivity,^[8–10] topological materials,^[11,11–13] spin dynamics,^[14–16] molecular dynamics,^[17–19] and electron beam manipulation.^[20–22] Although significant progress has been made in THz source development, efficiently generating and broadband detecting THz radiation remain key challenges that must be addressed for further advancements in THz technology.

Current THz source technologies include photoconductive antenna,^[23,24] nonlinear optical crystals,^[25,25,27] and plasma-based methods.^[28–31] These sources are advancing toward broadband, integration, and higher energy outputs. Recently, spintronic THz emitter with higher integration and broader frequency ranges, as well as robust structures with MV-level outputs, are emerging as a key trend.^[32–35] However,

challenges like low efficiency and low laser damage threshold remain.

Crystalline quartz is a hard material with unique symmetry and excellent optical properties, widely used in photonics and nonlinear optics.^[36] In the THz frequency domain, quartz is a common substrate material.^[37] Recently, quartz has gained significant attention for its remarkable detection capabilities.^[38] It can be used to determine the full polarization state of the strong THz pulses^[39] and THz magneto-optical sampling.^[40] As the trigonal lattice structure of quartz lacks centro-symmetry, several research groups have reported observing higher-order nonlinear optical effects in quartz.^[41,42] Our previous study demonstrated both THz Pockels and Kerr effects in alpha-quartz.^[43] In particular, quartz has a higher laser damage threshold and better thermal stability than conventional materials like ZnTe and GaP,^[44,45] making it a promising candidate for high-power THz sources.^[46] However, very little is known about the THz radiation mechanism and properties of crystalline quartz plate.

In this paper, we systematically investigate the twin pulsed THz emission characteristics of crystalline quartz under femtosecond laser excitation, in a transmission configuration. By measuring the time-separated THz emission signals with opposite polarity, we reveal the mechanism of the THz

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generation from crystalline quartz. These two pulses are attributed to the forced and free THz pulses, respectively. Furthermore, the dependences of the THz radiation on azimuthal angle and polarization angle of the pump laser beam are measured to characterize the THz generation spectral signatures.

2. Experimental setup

A THz time-domain emission spectroscopy system in transmission geometry was utilized to investigate the THz emission from crystalline quartz at room temperature. The laser beam in this experiment, from a Spitfire Ace titanium sapphire laser (Spectra-Physics) with 1 kHz repetition rate, 56 fs pulse duration, 5 mJ single pulse energy and 800-nm central wavelength, was split into a pump beam and a probe beam. The pump beam (2.5 mm in diameter) excited the sample. The pump beam was incident along the c -axis of z -cut crystalline quartz, as illustrated in Fig. 1(a). The polarization states of the pump beam can be adjusted with a half-wave plate. The emitted THz radiation was collected, collimated, and focused onto a 1 mm thick ZnTe (110) crystal using a pair

of off-axis parabolic mirrors (OAPM). The weak probe beam was focused onto the ZnTe crystal, spatially and temporally overlapping with the generated THz pulses. The probe beam passed through a quarter-wave plate (QWP) and a Wollaston prism (WP) before being detected by a couple of balanced photodetectors (PDs). The temporal signal was achieved by varying the time delay between the pump laser and the probe laser pulse. The measured EO sampling (EOS) pulse width was the convolution of THz radiation with detector response. A Teflon filter was used to block the residual 800 nm pump beam, while the generated THz signal can pass through.

To investigate the thickness dependence of THz emission, measurements were performed on commercially acquired crystalline quartz plates with thicknesses of 0.1 mm, 0.2 mm, 0.5 mm, and 1 mm, which were cut along the (001) crystallographic direction perpendicular to the plane. The crystallinity of the quartz plate has been characterized using x-ray diffraction pattern, which exhibits a pronounced (0001) characteristic peak at $2\theta = 50.62^\circ$.^[42] The azimuthal angle of the quartz plate can be varied by rotating the sample along the Z direction.

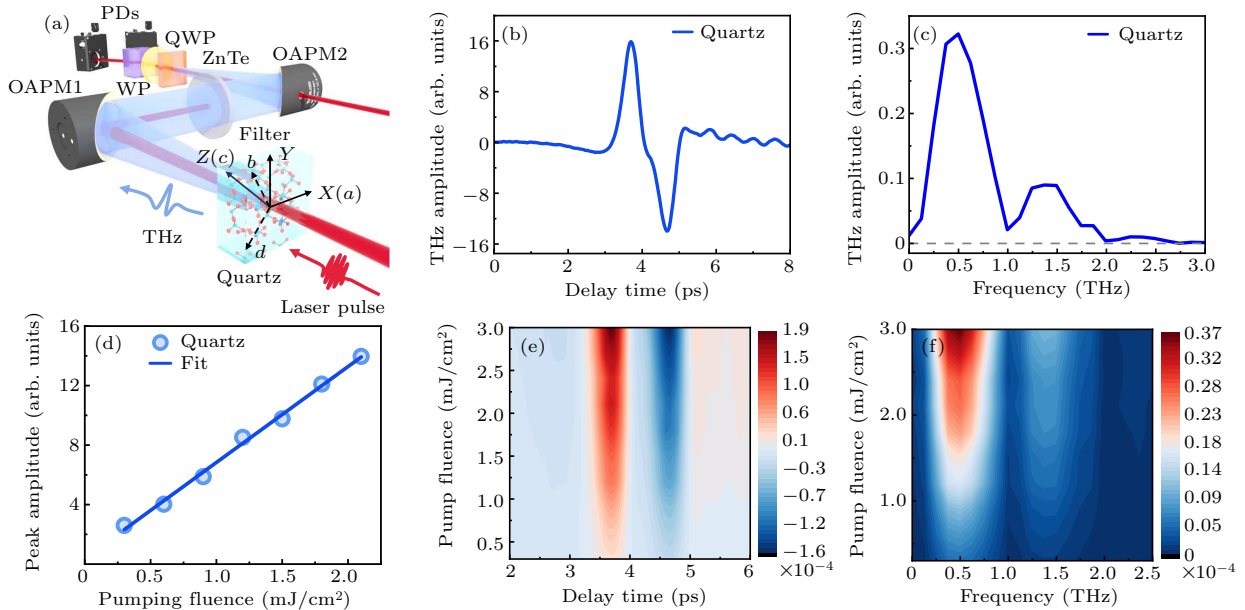


Fig. 1. (a) Schematic of THz emission setup. THz emission from quartz plate is measured in EOS configuration using a ZnTe detection crystal. (b) Time-domain THz emission waveform for a 0.5-mm-thick crystalline quartz plate. (c) Frequency spectrum after fast Fourier transformation. (d) Peak amplitude of THz signals as a function of pump fluence. (e) Contour map of the relationship between pump fluence and time-domain THz emission signals. (f) Corresponding spectra of time-domain traces in (e) after fast Fourier transformation.

3. Results and discussion

3.1. THz radiation

Figure 1(b) shows a typical time-domain THz emission signal from a 0.5 mm-thick crystalline quartz. The THz emission waveform exhibits a bipolar shape with almost equal amplitudes in both positive and negative peaks, which is different from the temporal shape of the rectification dipole. Fourier-transforming the time-domain signals yields the spec-

tral amplitude versus frequency, as shown in Fig. 1(c). The frequency spectrum covers a bandwidth from ~ 0.1 THz to 3.0 THz. Note that the spectral signatures, specifically the dips at ~ 1.0 THz and ~ 2.0 THz, result from the interference between the positive and negative peaks of the THz waveforms. As shown in Fig. 1(d), the THz peak amplitude increases linearly with the incident pump fluence ranging from 0.3 mJ/cm^2 to 2.1 mJ/cm^2 . This indicates that the THz emission is primarily a second-order nonlinear process.^[47] Figure 1(e) shows

the contour map of the relationship between pump fluence and THz signals in the time-domain. Figure 1(f) shows the contour map of the relationship between pump fluence and THz signals in the frequency-domain. It is clearly observed that as the pump fluence increases, the positions of the positive and negative peaks in the time domain do not exhibit any noticeable shift. Moreover, in the frequency domain, the dips remain unchanged with increasing pump fluence. It should be mentioned that the THz emission signal from fused silica glass can be negligible.

To better understand the mechanism of THz emission, we investigate the THz radiation of crystalline quartz with differ-

ent thicknesses. Figures 2(a) and 2(b) show the time-domain and frequency-domain characteristics of THz signals emitted from quartz plates with thicknesses of 0.1 mm, 0.2 mm, 0.5 mm, and 1.0 mm under a pump fluence of 1.5 mJ/cm^2 . Under the same excitation geometry, we compare the THz emission signals from quartz with ZnTe (black line), as shown in Fig. 2(a). The magnitude of THz emission from ZnTe is reduced by a factor of 100. As shown in Fig. 2(c), the positive peak amplitude from crystalline quartz increases with thickness up to $L = 0.5 \text{ mm}$, then decreases as thickness increases to 1.0 mm. The absolute values of the negative peak amplitude exhibit a similar dependence on the thickness.

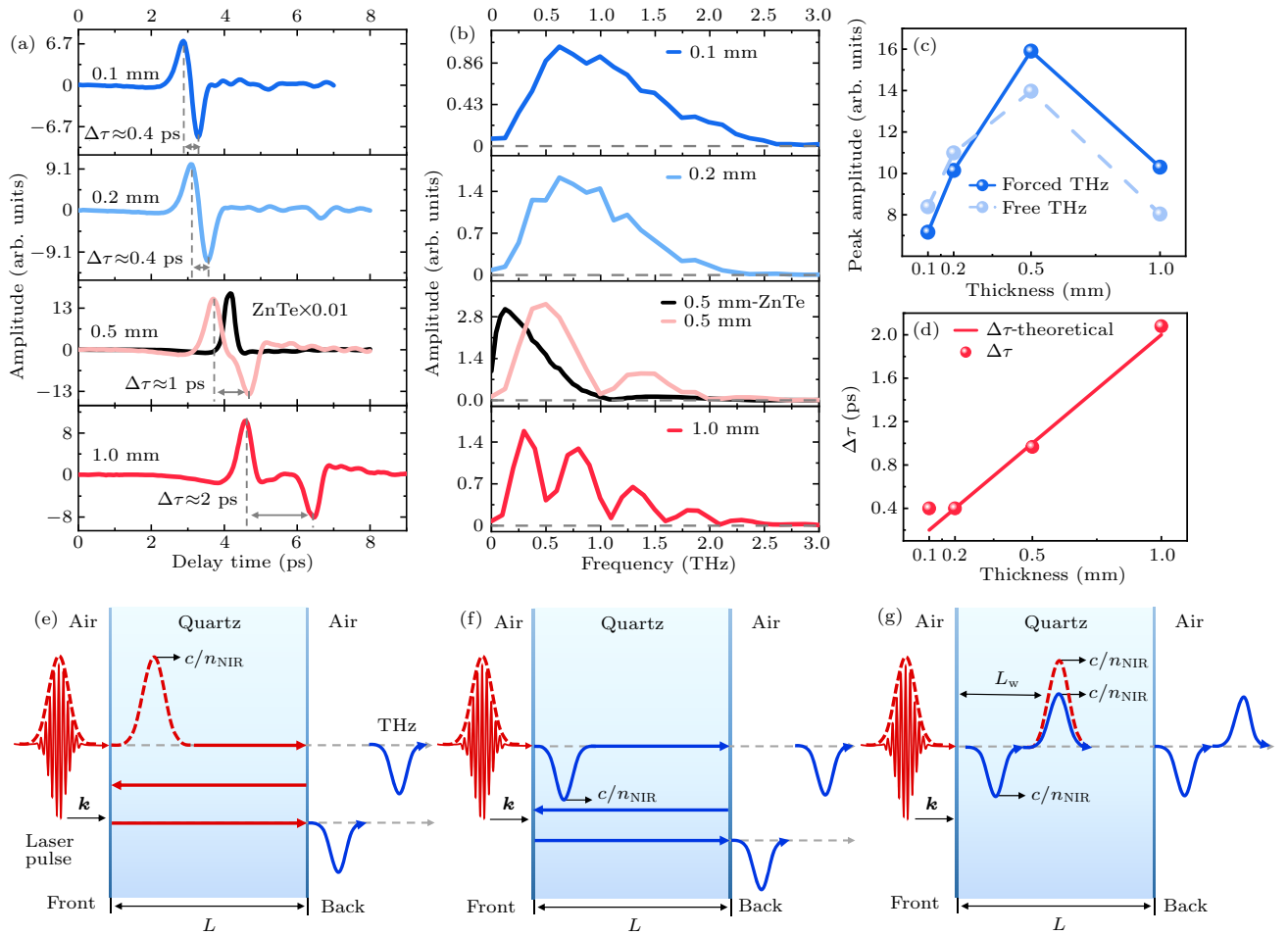


Fig. 2. (a) THz emission signals in the time domain obtained from crystalline quartz plates with varying thicknesses. (b) The corresponding frequency-domain spectra obtained after fast Fourier transformation. Dependence of (c) THz peak amplitude and (d) separation time on the sample's thickness. Schematic illustrations of (e) the laser pulses and (f) THz radiations inside the quartz plates. The sample's interfaces are labeled as front and back sides. (g) Both forced and free THz radiations are emitted near the front interface. The red and blue arrows show the propagations of NIR and THz pulses, respectively.

A time separation $\Delta\tau$ of $\sim 1 \text{ ps}$ is observed between the positive and negative peaks when the thickness of crystalline quartz is $L = 0.5 \text{ mm}$. When L decreases to 0.2 mm, $\Delta\tau$ reduces to $\sim 0.4 \text{ ps}$ and remains unchanged at $L = 0.1 \text{ mm}$. At $L = 1.0 \text{ mm}$, two peaks separate more clearly, with $\Delta\tau = 2 \text{ ps}$. This confirms that the bipolar waveform arises from the combination of two unipolar THz waves. We analyze the propagation and multi-reflection of near-infrared (NIR) pump light and THz waves within the quartz. As shown in Fig. 2(e), the NIR

pump pulse is reflected multiple times within the plate, resulting in a round trip delay of $\Delta\tau = 2n_{\text{NIR}}L/c$, with c the speed of light in vacuum, and refractive index $n_{\text{NIR}} \approx 1.55$ for quartz in the NIR range.^[48] At $L = 1 \text{ mm}$, $\Delta\tau \approx 10.3 \text{ ps}$, which is longer than the measured $\Delta\tau \approx 2.0 \text{ ps}$. As shown in Fig. 2(f), the emitted THz pulse is reflected multiple times inside the plate. At $L = 1 \text{ mm}$, $\Delta\tau = 2n_{\text{THz}}L/c \approx 14.4 \text{ ps}$, where the refractive index of crystalline quartz at THz frequency is measured by THz-time-domain spectroscopy to be $n_{\text{THz}} \approx 2.16$.^[42,43,49] It

is still longer than the measured $\Delta\tau \approx 2.0$ ps.

3.2. Primary mechanism

Optical rectification (OR) is a nonlinear process where an intense laser pulse generates low-frequency charge motion in a nonlinear medium, emitting THz radiation. The pump pulse induces nonlinear polarization $P(t)$ in the electrooptical material. The THz electric field is proportional to the second-order derivative of $P(t)$. Because of the presence of an optical pump pulse within the sample plate, a theoretical model is presented to describe the generation of both forced and free THz pulses via linear Maxwell's equation.^[50,51] The THz forced pulse propagates with a fixed velocity equal to the velocity of the optical pulses, unlike the case for a THz free pulse. As shown in Fig. 2(g), the emitted THz radiation, generated from the sample's front side, transmits through the plate and radiates out from the back side of the quartz plate. The nonlinear quartz plate is characterized by n_{THz} and n_{NIR} . In order to obtain an analytical result for the THz fields, the THz absorption and dispersion are neglected. In addition, it is assumed that a femtosecond laser pulse is incident normally on the plate and its transverse size is much larger than the THz wavelength. The orientation of the nonlinear polarization $\mathbf{P}$ depends on the orientation of the crystallographic axes of the sample and the polarization of the pump pulse. We assume $P_y = P_z = 0$ and $P_x \propto d_{ij} \times I_{\text{opt}}$ with I_{opt} the optical intensity of the pump beam and d_{ij} the second-order nonlinear susceptibility tensor. Under the assumption of normal incidence and neglecting THz absorption and dispersion, the frequency-domain THz radiation field $\tilde{E}_x$ generated by OR can be expressed as

$$\frac{\partial^2 \tilde{E}_x}{\partial z^2} + \frac{\omega^2}{c^2} n^2(z) \tilde{E}_x = -\frac{4\pi\omega^2}{c^2} P_x \tilde{F}(\omega) e^{-i\omega z n_{\text{NIR}}/c}, \quad (1)$$

where $\tilde{F}(\omega) = (\tau/2\sqrt{\pi}) e^{-\omega^2 \tau^2/4}$ is the Fourier transform of the time-dependent envelope of the optical intensity, and τ is the pulse duration.

By solving Eq. (1), the THz radiation is generated by the moving polarization,^[51]

$$E_x(z, t) = -\frac{n_{\text{THz}} - n_{\text{NIR}}}{n_{\text{THz}} + n_0} A e^{-(t + \frac{zn_0}{c})^2/\tau^2}, \quad z < 0, \quad (2)$$

$$E_x(z, t) = A \left[\frac{n_{\text{NIR}} + n_0}{n_{\text{THz}} + n_0} e^{-(t - \frac{zn_{\text{THz}}}{c})^2/\tau^2} - e^{-(t - \frac{zn_{\text{NIR}}}{c})^2/\tau^2} \right], \quad z > 0, \quad (3)$$

where $A = 4\pi p_x / (n_{\text{THz}}^2 - n_{\text{NIR}}^2)$, n_0 is the refractive index of air in the THz frequency range. For Eq. (2), it indicates the backward propagating THz pulses into the air ($z < 0$) after the optical pulse enters the plate. In the plate, Eq. (3) indicates the generation of two THz pulses with the same shape, but different amplitudes and opposite signs. The first term in Eq. (3) represents the free THz pulse, while the second term represents the forced THz pulse. The ratio of amplitude of the free

THz to that of the forced THz pulse is $\frac{n_{\text{NIR}} + n_0}{n_{\text{THz}} + n_0} = 0.81$. The free THz pulse and the forced THz pulse in the plate propagate with different velocities determined by n_{THz} and n_{NIR} , respectively. In the superluminal case ($n_{\text{THz}} > n_{\text{NIR}}$), the free THz pulse travels slower than the forced one.

Near the front side ($z = 0$), the free and forced THz pulses overlap and compensate each other partially. Thus, a walk-off length can be defined as $L_w = \frac{c\tau_{\text{FWHM}}}{|n_{\text{THz}} - n_{\text{NIR}}|}$, at which the THz pulses become separated. L_w is distinguished from the coherence length.^[51,52] The coherence length is defined as the effective length over which both optical and THz pulses propagate with the same velocity in the crystal, until they reach 180° phase difference. If we neglect dispersion in the optical spectral range, the coherence length can be expressed as $l_c = \frac{\pi c}{\omega_{\text{THz}} |n_{\text{NIR}} - n_{\text{THz}}|}$. In our case of crystalline quartz excited with pump pulse ($\tau_{\text{FWHM}} = 2\sqrt{\ln 2} \tau = 56$ fs), $n_{\text{NIR}} \approx 1.55$, $n_{\text{THz}} \approx 2.16$, we have $L_w \approx 27.5$ μm . Therefore, for a typical plate with $L > 100$ μm , the free and forced THz pulses arrive at the back side separately. In the process of propagation, the free and forced THz pulses become separated, $\Delta\tau = (n_{\text{THz}} - n_{\text{NIR}})L/c$. For quartz plates with different thicknesses, $\Delta\tau(0.1 \text{ mm}) \approx 0.2$ ps, $\Delta\tau(0.2 \text{ mm}) \approx 0.4$ ps, $\Delta\tau(0.5 \text{ mm}) \approx 1.0$ ps and $\Delta\tau(1.0 \text{ mm}) \approx 2.0$ ps. The calculated results are in agreement with the measured data, as shown in Fig. 2(d). While, we note that because the FWHM of a single THz pulse is approximately 0.4 ps, the $\Delta\tau$ values for 0.1 mm and 0.2 mm quartz are difficult to distinguish, both being close to 0.4 ps. Our analysis allows one to avoid using the explanation of the second THz radiation at the back side.^[53,54]

3.3. Analysis of the OR effect

To further confirm THz radiation from the OR effect, we investigate the azimuthal angle (φ) and polarization angle (α) dependences of the THz emission from the 0.5 mm thick crystalline quartz, which exhibits the highest THz generation. Figure 3(a) shows the time-domain signals and Fig. 3(b) presents the corresponding frequency-domain signals from quartz with different azimuthal angles. In the experiments, the pump laser beam is p-polarized. We find that the polarity of both free and forced THz pulses reverses every 60° of φ . Figure 3(c) shows the dependence of forced and free THz peak values as a function of φ , which presents perfect threefold symmetry. As shown in Fig. 3(d), the ratio of peak value of the free THz to that of the forced THz pulse presents an average value of 0.87 ± 0.13 , demonstrating good agreement with the theoretical value and verifying the validity of our proposed mechanism. In addition, a small phase shift $\sim 2^\circ$ is observed between the forced and free THz pulses.

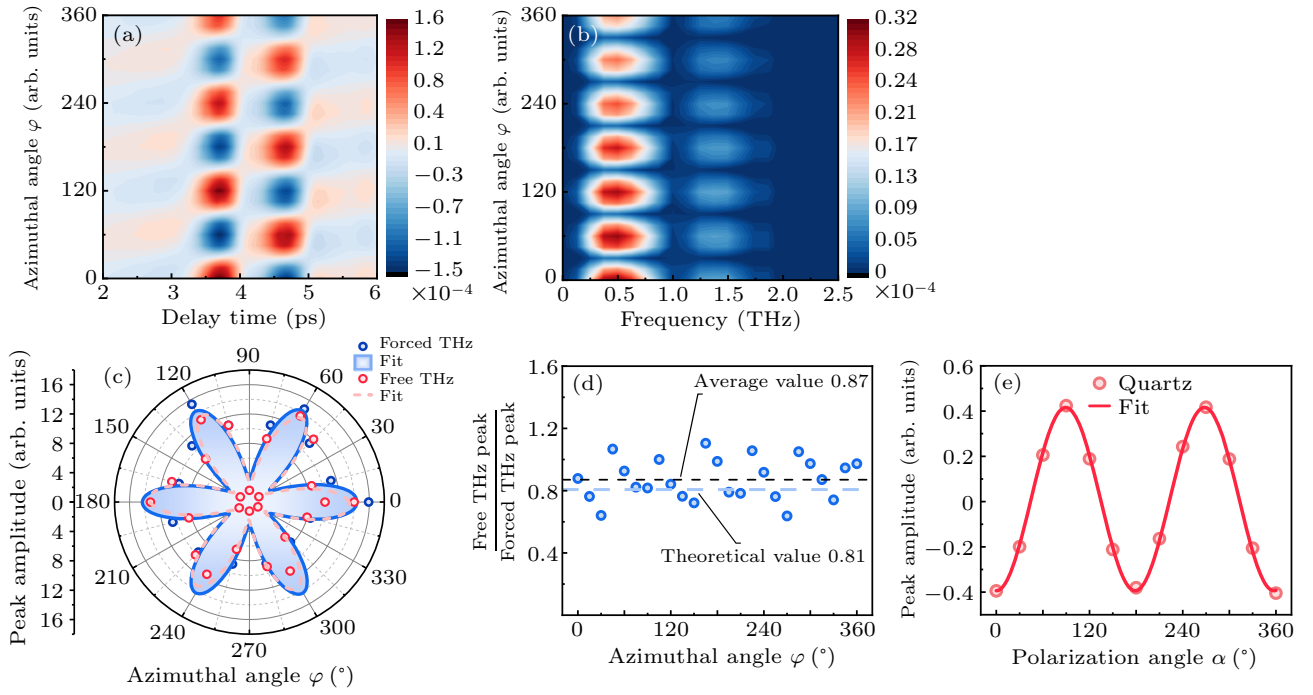


Fig. 3. (a) Contour map shows the relationship between the crystalline quartz azimuth and THz emission signals in time domain. (b) The corresponding THz spectra after Fourier transform. (c) Polar plots of free and forced THz peak amplitudes as functions of azimuth angle. (d) The ratio of peak value of the free THz pulse to that of the forced THz pulse as a function of azimuth angle φ . (e) Dependence of the THz peak amplitude on the polarization angle α of the pump pulse.

Crystalline quartz belongs to the D_{32} point group. For this symmetry group, the z -cut quartz lacks centrosymmetry.^[55] The crystal exhibits three-fold rotational symmetry along the c -axis and two-fold symmetry around the a -axis. It has two non-zero tensor elements of the effective second-order susceptibility, $d_{11} = 1.41$, and $d_{14} = 0.6$.^[56] The crystalline quartz satisfies type II phase matching, which depends on the internal phase matching angle θ and the azimuthal angle φ . The effective second-order susceptibility $d_{\text{eff}} = d_{11} \cos \theta \cos 3\varphi$.^[57] In our experiments, the optical pump beam co-propagates with the THz wave along the c -axis of the quartz plate, $\theta = 0^\circ$. Thus, the amplitude of the THz radiation can be written as $E_{\text{THz}}(\varphi) \propto d_{11} \cos 3\varphi \times I_{\text{opt}}$. This means the THz peak amplitude of quartz shows six maxima with respect to φ , consistent with our experimental results.

In addition, the THz amplitude as a function of the incident polarization angle of the linearly polarized pump pulse can be expressed as $E_{\text{THz}}(\alpha) \propto d_{11} \cos(2\alpha) \times I_{\text{opt}}$, where α characterizes the linear-polarization state with respect to the p-polarized pump pulse ($\alpha = 0^\circ$). It implies that the THz amplitude would appear with a period of 180° with respect to α , which agrees well with our experimental results in Fig. 3(e). The THz amplitude is symmetrical about the zero point, which confirms the THz emission from quartz is barely from the OR effect. In contrast, the THz emission of GaTe and diamond has the contributions from photocurrent within the surface and photo-drag-induced nonlinear photocurrent, respectively.^[58,59]

4. Conclusion

We have characterized the THz emission in z -cut crystalline quartz plates by using femtosecond laser pumping. Twin THz pulses emitted in the quartz plate are interpreted as the free and forced pulses based on Maxwell's equations. In addition, the THz radiation waveforms under different azimuthal angles of the sample and different polarization states demonstrate a second-order nonlinear THz generation process. The theoretical analysis of the interaction between the incident laser and the material is in good agreement with our experimental results. This investigation not only elucidates the mechanism of THz generation in crystalline quartz, but also paves the way for the development of electro-optic media for strong-field THz devices.

Acknowledgements

Project supported by the National Key Research and Development Program of China (Grant No. 2023YFF0719200), the National Natural Science Foundation of China (Grant Nos. 62322115, U24A20226, 61988102, and 62435010), 111 Project (Grant No. D18014), Science and Technology Commission of Shanghai Municipality (Grant Nos. 22JC1400200 and 21S31907400).

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