Narrow-band and tunable intense terahertz pulses for mode-selective coherent phonon excitation

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ABSTRACT

We generate frequency-tunable narrow-band intense fields in the terahertz (THz) range by optical rectification of a temporally modulated near-infrared laser pumping a nonlinear organic crystal. Carrier-frequency tunability between 0.5 and 6.5 THz is achieved by changing the modulation period of the laser pump. This tunable narrow-band THz source allows the selective coherent excitation of adjacent vibrational modes, which are demonstrated for two phonons with a frequency offset of 0.8 THz in single-crystal $\text{SrCu}_2(\text{BO}_3)_2$. Our compact and scalable source enables an effective approach for the advanced manipulation of low-energy collective modes in condensed matter and has the potential to reveal the coupling of specific lattice vibrations with other degrees of freedom.

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The generation of coherent phonons using light is an emerging tool to explore the dynamics of collective lattice vibrations, and it is expected to play a major role in the ultrafast control of functional electronic, magnetic, and structural properties of solids.^{1–4} In conventional coherent phonon spectroscopy, sub-picosecond visible or near-infrared lasers trigger coherent lattice displacements via Raman scattering.⁵ In this second-order process, which relies on excitation of the electronic system by high-energy photons,^{6,7} the amplitude of the coherent phonons is limited by the magnitude of the electron-lattice interaction. Moreover, the laser-driven generation of coherent phonons involves an excess of photo-induced hot electrons and, therefore, offers a restricted scalability associated with the large increase in sample temperature and ultimately with the laser-induced damage.

An efficient approach to overcome these limitations and to access phonon-mediated dynamics is based on the resonant excitation of infrared-active phonon modes using low-frequency light pulses. This radiation has the potential to generate the large-amplitude lattice displacements required to explore phonon-driven processes. Because the optical phonon frequencies in solids lie primarily in the spectral region between 1 and 10 THz, these experiments demand intense and phase-stable optical pulses at terahertz (THz) frequencies.^{8,9} The recent development of strong-field quasi single-cycle THz sources has enabled the investigation of phonon-driven phenomena and numerous other

coherent low-energy excitations, which were previously restricted. However, broadband THz sources drive multi-mode resonances, hindering the deep understanding of transient dynamics. An intense and narrow-band THz source tunable to the specific phonon resonance would greatly expand the understanding of phonon-mediated dynamics targeting distinct modes.

In this Letter, we present a scheme to generate intense narrow-band THz pulses by optical rectification of temporally shaped near-infrared laser pulses in a nonlinear organic salt DSTMS (4-N, N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate).¹⁰ We demonstrate the generation of narrow-band THz pulses with a bandwidth less than 1 THz and carrier frequency tunability between 0.5 THz and 6.5 THz. The continuous tunability of intense THz pulses represents a key development for selective phonon excitation targeting distinct infrared-active modes. As an example, we demonstrate the selective excitation of the adjacent phonon lines in single crystal $\text{SrCu}_2(\text{BO}_3)_2$.

The scheme we present has numerous advantages such as high fields and, moreover, fills the spectral gap of the existing laser-based narrow-band THz sources, such as the difference-frequency generation (DFG) in organic crystals,¹¹ which is suitable for higher frequencies (4–18 THz), or optical rectification by temporal modulated pulses in LiNbO_3 ^{12,13} and HMQ-TMS,¹⁴ which can cover lower frequencies of 0.3–1.3 THz and 0.3–0.8 THz, respectively. Our source is tunable

over a spectral range, 0.5–6.5 THz, comparable with the one demonstrated using 10 Hz Cr:forsterite to pump an organic crystal,^{15,16} but relies on a more commercially available optical-parametric amplifier (OPA) pump, which has the advantages of a higher repetition rate and a tunable wavelength for optimal pumping of different organic crystals. The higher repetition rate achievable with an OPA and the high field demonstrated here turn into a higher signal-to-noise ratio, which is crucial for future spectroscopic applications.

In the experiment, ultrashort near-infrared (NIR) laser pulses at a wavelength of 1505 nm, energy up to 2.6 mJ, and a pulse duration of approximately 60 fs are generated in a three-stage OPA (Light Conversion) driven by a terawatt-class Ti:sapphire laser front end,¹⁷ shown in Fig. 1(a). The OPA pulses are temporally stretched by two pairs of high-efficiency transmission gratings (996.4 grooves/mm) so that the instantaneous laser frequency is chirped linearly in time across the pulse duration. The distance between the first and the second grating pair is $d = 1.75$ mm. The temporal modulation is achieved by splitting, delaying, and recombining two chirped replicas in a Mach-Zehnder interferometer (MZI). For a linearly chirped optical pulse with a Gaussian envelope,¹⁸ the electric field can be described as

$E(t) = E_0 \exp(-\alpha t^2) \exp(i\omega_0 t + ibt^2)$, where E_0 is the electric field amplitude, $\omega_0/(2\pi)$ is the laser carrier frequency, b is the chirp rate, and $t_c = 2\sqrt{2 \ln 2/\alpha}$ is the FWHM duration of the chirped pulse, which has been estimated to be approximately 1.55 ps from propagation simulation using the ZEMAX optical design code. The resulting temporal intensity from the superposition of these two time-shifted replica pulses by the MZI is:¹⁸

$$I(t, \tau) = |E(t) + E(t + \tau)|^2 = I_1(t) + I_2(t + \tau) + 2\sqrt{I_1(t)I_2(t + \tau)} \cos(\omega_0 \tau + b\tau^2 + 2b\tau t). \quad (1)$$

The time delay τ of the two pulses produces a Gaussian-enveloped, periodically modulated intensity with a beat frequency $\Omega = b\tau/\pi$, which is proportional to the chirp rate b . The temporally modulated pulse exhibits a comb-like structure in the frequency domain, where Ω corresponds to the comb spacing. The time delay τ between the two arms of the MZI can be changed to easily achieve the continuous tunability of the modulation frequency. Figure 1(b) shows the spectrum of the laser pulse measured by a fiber-coupled spectrometer (NIRquest OceanOptics, resolution 3.3 nm). The beat frequency is $\Omega = 1.8$ THz for $\tau = 0.637$ ps. The temporally modulated pulse generates narrow-band THz radiation by optical rectification in a 500 ± 15 μm thick DSTMS (Rainbow Photonics). Because the chirped pulses are relatively long (~ 1.5 ps), the optical rectification from the individual Gaussian envelope is suppressed. This is confirmed by the negligible THz emission when the two replicas have no temporal overlap. The optical rectification becomes efficient when the beat frequency Ω is within the THz emission bandwidth of the DSTMS. Behind the THz generation, three low-pass filters (two at 20 THz and one at 10 THz, made by QMC Instruments) are used to block the residual NIR beam (the overall extinction ratio for the pump exceeds 10^5).

To reach intense fields, the THz beam is focused tightly by means of three off-axis parabolic mirrors (focal lengths of 1, 6, and 3 in.), achieving the spot size of around 1.5 times the diffraction limit. The temporal THz field is measured by electro-optic sampling (EOS) in 200 μm -thick GaP crystals using a 50 fs, 800 nm 45 μm low energy gating pulse obtained from the front-end Ti:sapphire laser.

Figure 2(a) shows the multi-cycle THz electric field generated by the modulated NIR pulse of Fig. 1(b). The spectral intensity, in Fig. 2(b), shows a Gaussian-like spectrum with its centroid at the beat frequency $\Omega = 1.8$ THz and a FWHM bandwidth of 0.47 THz.

FIG. 1. Narrow-band THz generation setup. The output of an optical parametric amplifier (OPA) driven by a Ti:sapphire laser is chirped linearly by four high-efficiency transmission gratings (G1–G4). The pulses are split and delayed in the Mach-Zehnder interferometer (MZI) creating Gaussian-enveloped temporally modulated pulses at beat frequency Ω , which is selected by varying the delay τ . The temporally modulated pulses drive the optical rectification in DSTMS, generating narrow-band THz pulses at frequency Ω . The generated THz fields are detected by EOS in a 200 μm thick GaP crystal. WP: Wollaston prism, QWP: quarter-wave plate, and PM1-PM4: parabolic mirrors. (b) Pump spectrum (blue curve) after the MZI for $\tau = 0.637$ corresponding to a beating frequency of $\Omega = 1.8$ THz. Reference spectrum (red dotted curve) in the absence of interference. The corresponding spectral modulation, calculated by removing the reference, is shown in the inset.

FIG. 2. (a) Electric field of the multi-cycle THz pulse generated for $\tau = 0.637$ ps by optical rectification in 500 μm thick DSTMS from the temporally modulated beam with the spectrum reported in Fig. 1(b). (b) Corresponding power spectrum. The THz emission by optical rectification is centered at the beat frequency $\Omega = 1.8$ THz.

As observed in Fig. 2(a), the THz pulse shows a linear frequency chirp, which is due to the undesired third-order dispersion (TOD) introduced by the transmission gratings (see the [supplementary material](#) for the experimental evaluation and the analytical description).

The peak electric field $E_{peak} = \sqrt{z_0 E_p 4\sqrt{\ln 2} / (\pi\sqrt{\pi} w)^2 \tau_{FWHM}}$ = 110 ± 5 kV/cm was evaluated experimentally¹⁹ by measuring (i) the THz pulse energy, $E_p = 0.15 \pm 0.01$ μ J, with a calibrated Golay cell; (ii) the waist size $w = 390$ μ m, with a micro-bolometric THz camera (NEC IRV-T0830); (iii) the FWHM pulse duration, $t_{FWHM}^{THz} = 1.85$ ps, from the EOS time-trace. The stability of the source against thermal and mechanical drifts over time is confirmed by repeatable EOS traces and modulated laser spectrum at the MZI output.

By changing the delay τ , the THz center frequency can be continuously tuned over the emission band of the DSTMS, as shown in Fig. 3. Both the number of temporal oscillations and the THz carrier frequency increase with τ [Fig. 3(a)]. The corresponding spectra, shown in Fig. 3(b), indicate that the carrier frequency can be adjusted continuously between 0.5 and 6.5 THz (i.e., over 3.7 octaves). The THz center frequency depends linearly on τ , as shown in Fig. 3(c), with a slope of 3.3 THz/ps. Because the THz center frequency corresponds to the beat frequency, $\Omega = b\tau/\pi$, we can estimate a chirp rate of $|b| = 10.3$ THz/ps. The corresponding FWHM pulse duration of the chirped NIR pulse²⁰ can be estimated using the approximated expression $t_c = \Delta\omega/(2b) = 2.1$ ps (with $\Delta\omega = 2\pi \cdot 7$ THz the measured FWHM spectral bandwidth), which agrees qualitatively with the values 1.5 ps calculated from the ZEMAX simulation. The bandwidth of

the THz pulses generated at various τ , shown in the inset of Fig. 3(c), is lower than 0.5 THz for delays $\tau < 0.6$ ps, which corresponds to a center frequency below 2 THz. The smaller bandwidth at lower τ may be ascribed to the strong absorption in DSTMS around 1 THz. The bandwidth and the number of cycles of the THz radiation can be controlled by the chirp-rate changing of the grating spacing. For the present measurements, the grating spacing is kept constant. However, the presence of the THz frequency chirp introduced by the TOD (see the [supplementary material](#)) prevents the generation of ultra narrow-band THz pulses due to the spectro-temporal dispersion of the carrier frequency. To achieve narrower and unchirped THz generation, the laser temporal modulation can be implemented by grating stretcher,²¹ which allows us to directly control the laser temporal shape by optical masks or spatial light modulators in the Fourier plane. The highest THz electric field strength (410 kV/cm) is achieved at a frequency around 3.8 THz [Fig. 3(d)]. For this frequency, the measured THz energy is 0.5 μ J. As expected, the frequency-scaling reproduces the spectral features of THz generated by a transform-limited pulse, as shown by the red curve in Fig. 1(d). The inset of Fig. 3(d) shows the focal spot size at different THz carrier frequencies measured by the THz camera: the spot size scales as inverse of the THz carrier frequency. In Table I, we report the main parameters of the present narrow-band and tunable THz source compared to previous similar setups.

Our narrow-band THz source is well suited for the selective control of low-energy collective modes. In order to demonstrate its versatility, we perform a resonant coherent phonon spectroscopy experiment on single crystal $\text{SrCu}_2(\text{BO}_3)_2$. This material has attracted considerable attention in the field of quantum magnetism,²² and for our purpose, it has very sharp phonon modes at low temperature, which lie in the spectral range of our THz source. The sample, grown using the floating zone method, is 120 μ m thick in the c direction and has an area of ~ 4 mm² in the a “b” plane²³ [Fig. 4(a)]. The narrow-band THz pump pulse propagates along the c -axis with the electric field on the a “b” plane. In this configuration, $\text{SrCu}_2(\text{BO}_3)_2$ shows 3 non-fully symmetric E -modes²⁴ located in our THz pump frequency region: $\nu_1 = 3.8$ THz, $\nu_2 = 4.3$ THz, and $\nu_3 = 4.6$ THz. Because the E -symmetric modes are both IR-active, with the dipole moment in the a “b” plane, and Raman-active, they allow for a scheme of direct excitation at the Γ -point with THz pulse and femtosecond optical detection.^{25,26} By measuring the anisotropic transmission changes, $\Delta T/T$, at different delays with respect to the narrow-band THz pump, we detect the changes in susceptibility associated with the coherent atomic displacements related to E -modes. Similar to the EOS setup [Fig. 1(a)], the detected anisotropic transmission change, $\Delta T/T = (T_1 - T_2)/T$, is proportional to the orthogonal polarization intensities of the probe, I_1 and I_2 , shown in Fig. 4(a). These are separated by the Wollaston prism and detected by balanced photodiodes. T is the probe transmittance without the THz pump.

The temporal evolution of the measured $\Delta T/T$ is depicted in Fig. 4(b). When the carrier frequency of the THz pump is tuned around 4 THz, we observe large-amplitude coherent oscillations from resonantly driven phonon modes. Because at low temperature $\text{SrCu}_2(\text{BO}_3)_2$ is a Mott insulator and the pump photon energy is far below the optical gap $\Delta_0 = 0.7$ eV,²⁴ step-like functions or decay features related to excitation and relaxation of the electronic systems are not observed in the dynamics. Figure 4(c) shows that the spectral

FIG. 3. (a) Multi-cycle THz waveforms and (b) corresponding spectra obtained by tuning the delay τ . (c) Center frequency of the THz pulse shown as a function of τ . The gray solid line is a linear fit with a slope of 3.3 THz/ps. The inset shows the bandwidth of the narrow-band THz pulses. (d) Peak field of the multi-cycle THz pulses as a function of the center frequency (blue dots) compared to the spectral amplitude of the broadband THz pulse generated with a 60 fs NIR pulse in the same crystal (red curve). Inset: FWHM spot size of the THz beam at focus (red dots), which scales with the inverse of THz carrier-frequency. The gray solid line is the corresponding fit.

TABLE I. Parameters of the intense narrow-band and tunable laser-based terahertz sources.

Method	Tuning range (THz)	Bandwidth (THz)	Max THz energy (μJ)	Max strength (MV/cm)	Nonlinear crystal
Chirp + MZI (this work)	0.5–6.5	< 0.5	0.5	0.4	DSTMS
MTI + compressor ¹⁵	0.25–3	< 0.25	6.7	2.3	OH1
MTI + compressor (10 Hz) ¹⁶	0.7–7	< 0.5	20	8	DSTMS
Chirp + etalon ¹³	0.3–1.3	< 0.1	1.5	...	LiNbO ₃
Chirp+etalon ¹⁴	0.3–0.8	< 0.1	0.08	...	OHQ-TMS
DFG ¹¹	4–18	< 1	1.9	3.7	DSTMS

amplitude of the measured $\Delta T/T$ displays strong and narrow peaks at 3.8 THz and 4.6 THz.

To understand the observed dynamics, we model the coherent lattice displacement as a sum of damped harmonic oscillators driven by the THz electric field of the pump, as shown by the dotted black curve in Fig. 4(c). The equation of motion for normal coordinate $Q_i(t)$ along the displacement vector of the i th phonon mode is²⁵

$$\frac{d^2 Q_i(t)}{dt^2} + 2\gamma_i \frac{dQ_i(t)}{dt} + \omega_i^2 Q_i(t) = \frac{e_i^*}{m_i^*} E_{\text{THz}}^{\text{eff}}(t), \quad (2)$$

where $\omega_i = 2\pi\nu_i$ and γ_i are the phonon angular eigenfrequencies and the damping rate, while e_i^* and m_i^* are the effective charge and the reduced mass of the mode. The effective driving electric field $E_{\text{THz}}^{\text{eff}}(t)$ inside the sample corresponds to $E_{\text{THz}}^{\text{eff}}(\nu) = t(\nu)E_{\text{THz}}(\nu)$ in the frequency domain, where $E_{\text{THz}}(\nu)$ is the Fourier transform of the pump electric field, $t(\nu) = 2/(1 + \tilde{n}(\nu))$ is the THz transmission amplitude, and $\tilde{n}(\nu)$ is the complex refractive index, which is strongly frequency-dependent at the IR-active lattice resonances. The refractive index is derived from the complex dielectric function²⁴ $\tilde{\epsilon} = \epsilon_\infty + \sum_i \omega_{p,i}^2 / (\omega_i^2 - \omega^2 - i\gamma_i\omega)$, where $\omega_{p,i}$ is the plasma frequency and γ_i the damping rate. The anisotropic transmission change associated with the atomic coordinate $Q_i(t)$ is connected to the Raman tensor R_{kj}^i of

FIG. 4. (a) Lattice structure of SrCu₂(BO₃)₂ in the a'b' plane and experimental geometry. The THz pump electric field is polarized along the vertical direction. (b) Evolution of anisotropic transmission changes $\Delta T/T$ caused by THz excitation of the E -symmetric phonon modes. (c) Spectral amplitude obtained by Fourier transformation (red curve) and from the model of Lorentz oscillators given in Eq. (2) (black dotted curve). The sample temperature is 4 K.

the i th E -symmetry mode by $\Delta T/T \propto R_{kj}^i Q_i$. By using the parameters for the three phonon modes taken from Ref. 24, $\omega_{p,1} = 125 \text{ cm}^{-1}$ (3.8 THz), $\omega_{p,2} = 141 \text{ cm}^{-1}$ (4.3 THz), $\omega_{p,3} = 157 \text{ cm}^{-1}$ (4.7 THz), $\gamma_1 = 1.0 \text{ cm}^{-1}$, $\gamma_2 = 1.8 \text{ cm}^{-1}$, and $\gamma_3 = 0.7$, we calculate the transmission changes in the frequency domain by using Eq. (2) and the driving THz field measured by EOS. The only free parameters are the relative magnitudes of the peaks ($A_{E_1}, A_{E_2}, A_{E_3}$), which are linearly proportional to the dipole coupling terms e_i^*/m_i^* and to the amplitude of the Raman tensors R_{kj}^i of the modes. As shown in Fig. 4(c), the numerical calculation reproduces the experimental data and captures the relative peak ratios $A_{E_2}/A_{E_1} = 0.3$ and $A_{E_3}/A_{E_1} = 4$. This is expected from Raman spectroscopy because the intensities of E_1 and E_2 are larger than those of E_3 at room temperature²⁷ and conceivably also at low temperature. In the calculation, we assumed an equal coupling term e_i^*/m_i^* for the phonon modes because they have a comparable net dipole moment.²⁴ In Fig. 5, we show the THz-driven dynamics of $\Delta T/T$ as a function of

FIG. 5. (a) Dynamics of $\Delta T/T$ at different THz pump carrier frequencies: 1.4 THz, 3.2 THz, 4.6 THz, and 6.1 THz. (b) Corresponding spectral amplitude $\Delta T/T(\nu)$ for $t_{pp} > 4.2 \text{ ps}$ (when $E_{\text{THz}} = 0$) and related THz pump spectra (top panel). (c) Atomic displacement vectors projected in the a b plane for the observed infrared-active E_1 and E_3 phonon modes, from Ref. 24. (d) Normalized amplitude $\Delta T/T(\nu)$ for $t_{pp} > 4.2 \text{ ps}$ as a function of the carrier frequency of the THz pump. The 45° dashed line is used as a guide to the eye. Red and blue colors correspond to $\Delta T/T(\nu) = 1$ and $\Delta T/T(\nu) = 0$, respectively.

the pump carrier frequency. In particular, Fig. 5(a) displays the pump-probe time-traces for pump frequencies at 1.4 THz, 3.2 THz, 4.6 THz, and 6.1 THz. The corresponding Fourier transforms are shown in Fig. 5(b). For the off-resonant THz pump frequencies at 1.4 THz and 6.1 THz, no long-lived coherent oscillations are observed in the dynamics of $\Delta T/T$, while for pumping at 3.8 THz and 4.6 THz, the E_1 mode and E_3 resonances are excited selectively. The weak oscillations for the off-resonant pump are ascribed to the electro-optic response of $\text{SrCu}_2(\text{BO}_3)_2$. It is evident that the THz pump can target different atomic displacements associated with the E_1 and E_3 phonon modes, which are depicted in Fig. 5(c). In Fig. 5(d), we present the 2D map of the amplitude spectrum of $\Delta T/T$ as a function of the THz carrier frequency of the pump.

In summary, we generate intense and tunable narrow-band THz pulses by optical rectification in DSTMS of a modulated laser intensity profile. This source allows subterahertz linewidth and carrier-frequency tunability from 0.5 to 6.5 THz, which is not available with other high repetition-rate sources based on standard lasers. These characteristics are highly desirable for the selective excitation of IR-active modes and, in general, for advanced time-resolved spectroscopy. By using a coherent phonon detection scheme, we demonstrated that the narrow-band THz source can resonantly drive large-amplitude lattice displacements associated with non-fully symmetric modes in single-crystal $\text{SrCu}_2(\text{BO}_3)_2$. We found that the excited phonon spectrum is well reproduced by a model of Lorentz oscillators driven by multi-cycle narrow-band THz pulses. By tuning the carrier frequency of the THz stimulus, we can select a single phonon mode to drive. The compact source we present based on commercially available OPA is up-scalable to higher THz fields, offering viable approaches for the study of phonon dependent nonlinearities in condensed matter.

See the [supplementary material](#) for the experimental evaluation and the analytical description of the THz chirp.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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