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High-Brightness and Continuously Tunable Terahertz-Wave Generation

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Abstract

One of the interesting frequency regions lies in the “frequency gap” region between millimeter wave and infrared, terahertz (THz) wave. Although new methods for generating terahertz radiation have been developed, most sources cannot generate high-brightness (high-peak-power and narrow-linewidth) and continuously tunable terahertz waves. Here, we introduce the generation of high-brightness and continuously tunable terahertz waves using parametric wavelength conversion in a nonlinear crystal; this is brighter than many specialized sources such as far-infrared free-electron lasers. We revealed novel optical parametric wavelength conversion using stimulated Raman scattering in lithium niobate as a nonlinear crystal without stimulated Brillouin scattering using recently developed microchip Nd:YAG laser. Furthermore, we show how to optimize the tuning curve by controlling the pumping and seeding beam. These are very promising for extending applied research into the terahertz region, and we expect that this source will open up new research fields such as nonlinear optics in the terahertz region

Keywords: terahertz wave, nonlinear optics, parametric wavelength conversion

1. Introduction

Terahertz waves (wavelength, 30–3000 μm ; frequency, 10–0.1 THz) are important not only in the fields of basic science, such as molecular spectroscopy, molecular optics, plasma measurement, charged particle acceleration, and radio astronomy, but also in numerous applications, such as broadband wireless communication, nondestructive inspection, high precision radar,

and global environmental measurement, since they have higher directivity like infrared than microwaves and higher transmittances in the atmosphere and in soft materials like microwave than infrared. Therefore, high-peak-power, narrow-linewidth (high-brightness), and continuously tunable terahertz-wave sources that could be widely used in such applications are required. The terahertz (THz) region is relatively unexplored, because of the lack of the commercially available high-brightness and continuously tunable sources, high-sensitive and fast detectors, and optics, which has resulted in what is called the frequency gap [1–3]. Over the past two decades, there has been striking growth in the region of science and engineering, which has become a vibrant, international, cross disciplinary research activity [4]. Wavelength (frequency) conversion in nonlinear optical materials is an effective method for generating high-brightness and continuously tunable terahertz waves owing to the high conversion efficiency, bandwidth, wide tunability, and room temperature operation. A terahertz-wave source using parametric wavelength conversion based on lithium niobate (LiNbO_3) crystals was first proposed in 1960s [5, 6] and realized in the mid-1990s with the terahertz-wave parametric oscillator [7]. At that time, the tuning range and the observed maximum peak output power of terahertz wave were from 1.1 to 1.6 THz (270–184 μm) and several milliwatts, respectively. By using the current injection-seeded terahertz-wave parametric generator (is-TPG), the tuning range expanding from 0.39 to 5.0 THz (750–60 μm) and the peak output power exceeding 55 kW [8–11] were observed, representing an increase by 10 times and seven orders of magnitude, as shown in **Figure 1**. **Table 1** lists the characteristics of three typical intense terahertz-wave sources: our injection-seeded terahertz-wave parametric generator (is-TPG), well-known intense terahertz-wave sources, a far-infrared free-electron laser (FIR-FEL) [12], and terahertz-wave pulse generation through optical rectification using a tilted-pulsefront

Figure 1. Development of parametric sources using LiNbO_3 in our group. Points represent the peak output power during 1–3 THz in each paper.

	Peak power [kW]	Linewidth [GHz]	Tb [K]	Tuning range
is-TPG	~ 100	~ 4	~ 10^{18}	0.4 – 5
OR	~ 5000	~ 1500	~ 10^{17}	broadband
FIR-FEL	~ 2	~ 10	~ 10^{16}	1 - 5

Table 1. Characteristics of typical intense terahertz-wave sources: our injection-seeded terahertz-wave parametric generator (is-TPG), terahertz-wave pulse generation through optical rectification using a tilted-pulse-front excitation (OR), and a narrowband free-electron laser that works in the far-infrared region (FIR-FEL). The brightness temperature (T_b) is calculated as $k_b T_b = \text{Peak power} / [(M^2)^2 \times \text{linewidth}]$. OR generates broadband terahertz waves.

excitation (OR) [13]. The is-TPG is one of the brightest sources in the terahertz region with a wide tuning range. We explain in this chapter how the high-brightness terahertz waves are generated via acoustic phonons of a nonlinear lithium niobate (LiNbO_3) crystal.

2. Terahertz-wave parametric generation

When the intense laser beams pass through a nonlinear optical crystal, the transverse photon and phonon wave fields become coupled and behave as new mixed photon-phonon states called polaritons. Broadband terahertz-wave generation results from efficient parametric scattering of laser light via polaritons [5, 6]. The polaritons exhibit phonon-like behavior in the resonant frequency region (near the transverse optical (TO)-phonon frequency ω_{TO}); however, they behave like photons in the nonresonant low-frequency region, as shown in **Figure 2**. Generation of narrowband terahertz waves can be achieved by applying an optical resonator (in the case of the terahertz-wave parametric oscillator (TPO)) or injecting a “seed” (in the case of the injection-seeded terahertz-wave parametric generator (is-TPG)) for the idler wave [14]. The wide tunability is accomplished simply by changing the angle between the incident pumping beam and the resonator axis (in the case of TPOs) or the wavelength and axis of the

Figure 2. (Left) Dispersion relation of the polariton. (Right) Noncollinear phase-matching condition.

seeding beam satisfying the phase-matching condition (in the case of is-TPGs). In the parametric wavelength conversion process, a terahertz-wave signal photon and a near-infrared idler photon are created parametrically from a near-infrared pumping photon, according to the energy conservation law $\omega_p = \omega_T + \omega_i$ (where ω indicates frequency and p, T, and i denote the pumping, terahertz, and idler photons, respectively) and the momentum conservation law $k_p = k_i + k_T$ (noncollinear phase-matching condition). This condition leads to the angle-dispersive characteristics of the idler and terahertz waves. Thus, broadband terahertz waves can be generated depending on the phase-matching angle.

In this experiment, we used a magnesium oxide (MgO)-doped LiNbO₃ crystal. The large figure of merit ($\text{FOM} = 4d_{\text{eff}}^2/n_{\text{NIR}}^2 n_{\text{THz}}^2 \alpha_{\text{THz}}^{-2} \sim 10$, d_{eff} : the effective nonlinear coefficient, n_{NIR} and n_{THz} : the refraction indices in the near infrared and terahertz range, α_{THz} : the absorption coefficient for the terahertz wave) [13] of LiNbO₃ at room temperature makes this well-known nonlinear crystal ideal for such an application. The gain curve of the terahertz-wave parametric generation is determined by the parametric gain and absorption coefficients in the terahertz region. **Figure 3** shows the pumping intensity dependence (0.1, 0.2, 0.4, 0.8, 1.6, and 3.2 GW/cm²) of calculated gain curves [15]. As the pumping intensity increases, the gain coefficient also increases in whole frequency region, and the maximum value in the gain curve moves toward higher frequencies. All gain curves have a broad bandwidth, with a drop appearing at around 2.6 THz. This is because the low-frequency modes of doped MgO in the LiNbO₃ work as crystal lattice defects. Under the noncollinear phase-matching condition, the effective gain curve depends on both the intensity and the beam diameter of the pumping beam. **Figure 4** shows the pumping beam diameter dependence of calculated effective gain curves. When an Nd:YAG laser

Figure 3. Pumping intensity dependence of calculated gain coefficient using MgO:LiNbO₃ pumped by Nd:YAG laser, when the pumping intensities were 0.1, 0.2, 0.4, 0.8, 1.6, and 3.2 GW/cm².

Figure 4. Pumping diameter dependence of effective calculated gain coefficient, when the pumping beam diameters were 0.5, 1, 2, and 5 mm. The pumping intensity was 1 GW/cm².

($\lambda = 1064$ nm) is used to generate the pumping beam and an MgO:LiNbO₃ crystal is used as the nonlinear optical crystal, the effective gain coefficient is given [16–18] by

$$g_T = \alpha \left[\left(1 + 16 \cos \phi (g_0/\alpha)^2 \right)^{1/2} - 1 \right] / 2,$$

$$\alpha = \alpha_{crystal} + \alpha_{beam},$$

$$\alpha_{beam} \propto \sin \phi / r_p,$$

where $\alpha_{crystal}$ is the absorption coefficient of the terahertz wave in the MgO:LiNbO₃ crystal, ϕ is the phase-matching angle between the pumping beam and the terahertz wave, g_0 is the parametric gain under the low-loss limit, and r_p is the diameter of the pumping beam. When the pumping beam intensity is 1 GW/cm², all gain curves (pumping beam diameter 0.5, 1, 2, and 5 mm) have enough gain coefficient over a broad range extending from less than 1 THz to more than 3 THz. As the pumping beam diameter becomes larger, the gain coefficient also increases, and the maximum value of the gain curve moves toward lower frequencies. As a result, we can optimize the tuning curves by controlling these parameters.

When a high-intensity laser beam propagates through a nonlinear crystal, a number of non-linear processes occur, such as the following: second- or higher-harmonic generation (SHG or HHG); difference- or sum-frequency generation (DFG or SFG); optical parametric generation, amplification, or oscillation (OPG, OPA, or OPO); stimulated Raman or Brillouin scattering

(SRS or SBS); four-wave mixing; optical rectification (OR); multiphoton absorption; and the Kerr and Pockels effects. Of these, we revealed that the parametric wavelength conversion near the lattice resonance induced by SRS is significantly inhibited by SBS; however, this nonlinear process has long been ignored. In the previous research done by authors, the conversion efficiency in energy from an infrared pumping beam to a terahertz wave was less than 10^{-7} . It has long been thought that this is the limit of the conversion efficiency using parametric wavelength conversion using LiNbO_3 pumped by nanoseconds Nd:YAG lasers (duration, 10–25 ns) [14]. However, when a photon of the pumping beam (1064 nm) creates two photons (idler beam and terahertz wave (100–1000 μm), in principle, the conversion efficiency reaches 10^{-2} – 10^{-3} according to the Manley-Rowe relations because the wavelength of the terahertz wave is about 10^2 – 10^3 times longer wavelength than that of the pumping beam. In our experiment, an infrared pumping beam excites acoustic phonons in LiNbO_3 , and SRS of the pumping beam generates terahertz waves and an idler beams. We calculated both gain coefficient of the SRS and the SBS in the previous condition; the gain coefficient of SBS has 1000 times larger gain than that of the SRS [19–24]. Typically, the SBS gain reaches the steady state within 10 lifetimes of the acoustic phonon of crystal [25], within about 1.5 ns in LiNbO_3 [24]. For efficient wavelength conversion, the pulse width of the pumping beam should be enough less than this, but the pulse width limits the linewidth of the generated terahertz waves. By applying a single-mode oscillated microchip Nd:YAG laser [26] with a sub-nanosecond (several hundreds of picoseconds) “pulse gap” pulse width [27] as a pumping source, a high-efficiency and narrow-linewidth wavelength conversion can be performed by the SRS without the SBS. Additionally, when the intensity of the pumping beam is too high, second-order stoke (idler) beams can be generated, which do not contribute to the generation of terahertz waves as they undergo strong absorption. We thus precisely controlled both pumping and seeding intensity and diameter as well as the nonlinear crystal length.

3. Experiment

The experimental apparatus, shown in **Figure 5**, consists of a pumping source (pulsed, Nd:YAG laser), a seeding source (CW, external cavity diode laser (ECDL)), amplifiers (for both pumping and seeding beams), and the nonlinear crystal ($\text{MgO}:\text{LiNbO}_3$). The pumping source is a diode end-pumped microchip $\text{Nd}^{3+}:\text{YAG}$ laser passively Q-switched by $\text{Cr}^{4+}:\text{YAG}$ saturable absorber. This configuration enables the low-order axial and transverse mode laser oscillation, whose linewidth is below 0.009 nm. The laser delivers more than 1.4 MW peak power pulses (energy/pulse, > 0.6 mJ/pulse; duration, ~ 420 ps) at 100 Hz repetition rate with a M^2 factor of less than 1.1 [26]. The pumping beam is amplified by two amplifiers in double-pass configurations. Each amplifier has 0.7 at.% doped $\text{Nd}^{3+}:\text{YAG}$ with a length and diameter of 70 and 3 mm, transversely pumped by 200 W laser diodes (wavelength, 808 nm) in a threefold geometry. Amplified beam is extracted by a polarization beam splitter (PBS). The seeding beam from an ECDL is also amplified by an Yb -doped fiber amplifier. Owing to the grating and confocal arrangement, the noncollinear phase-matching condition is satisfied automatically depending on the wavelength of the seeding beam [28]. The diameter of both beams is the same on the

Figure 5. Experimental apparatus for an is-TPG.

crystal input surface. We used a 50-mm-long nonlinear MgO:LiNbO_3 crystal with antireflection coating for a pumping beam. A prism made by high-resistivity silicon placed on the output surface of the nonlinear crystal works as an efficient output coupler only for the terahertz waves to avoid the total internal reflection of the terahertz waves on the crystal output side surface. For an optimization of terahertz-wave emission, the pumping region within the nonlinear crystal must be as close as possible to the output surface, because of the large absorption coefficient of the MgO:LiNbO_3 crystal in the terahertz range ($10\text{--}100\text{ cm}^{-1}$). The distance between the output surface and the beam center was precisely adjusted to obtain a maximum terahertz-wave output, and it was approximately equal to the pumping beam radius. The terahertz-wave output extracted through the Si-prism coupler was collimated, focused, attenuated, modulated, and then measured using a calibrated pyroelectric detector covered by thick black polyethylene sheet. The temporal waveform and linewidth of the terahertz wave were measured by a Schottky barrier diode (SBD) and a pair of scanning metal mesh plates.

4. Result and discussion

Figure 6 shows the tuning curves of two is-TPGs fabricated using our design obtained by scanning the wavelength of seeding beam. When the pumping beam diameter and energy are 1.5 mm and 20 mJ/pulse, and the seeding beam power is 800 mW (continuous wave), the tunable range of the terahertz wave is 0.7–3 THz ($430\text{--}100\text{ }\mu\text{m}$). The maximum output peak power is more than 55 kW ($\text{BT} \sim 10^{18}\text{ K}$, brightness $\sim 0.2\text{ GW/sr}\cdot\text{cm}^2$) at around 1.8 THz. This source has a broad tuning range, with a flat region around 1.6–2.6 THz. The terahertz-wave output decreased in the low- and high-frequency regions (below 1.6 and above 2.6 THz) because of a low parametric gain and high absorption coefficient [29] in these regions, respectively. From the is-TPG, the pulsed terahertz waves are generated by 100 Hz (by 10 ms); however, the pyroelectric detector we used in the experiment only gives an average power. We therefore

Figure 6. The tuning curves of two is-TPGs with pumping beam diameters of 1.5 mm, represented by blue solid line and 2.2 mm by red one.

used an optical chopper to measure the average power. We estimated the energy/pulse from the calibrated average power, and the maximum energy/pulse was more than 5.5 $\mu\text{J}/\text{pulse}$. When the thick glass plate as an IR pass filter was inserted, the output signal from the detector completely disappeared. We measured the duration of generated terahertz wave around 100 ps by the SBD, corresponding to peak power of more than 55 kW at 1.8 THz. The conversion efficiency in energy from pumping beam to terahertz wave is about 10^{-4} in this case. The tuning curve for a 2.2-mm-diameter pumping and seeding beam is also shown in **Figure 6**. In that case, the tunable range is 0.39–2 THz (760–150 μm). The tuning curve has an extremely broad bandwidth, and the lowest frequency (longest wavelength) of 0.39 THz (760 μm) was also the lowest frequency (longest wavelength) achieved in our experiment. The maximum terahertz-wave output is more than 7 kW (energy, 0.7 $\mu\text{J}/\text{pulse}$; duration, 100 ps), which occurs near 330 μm (0.9 THz) when the pumping energy is 15 mJ/pulse and seeding power is 2400 mW. The conversion efficiency is about 10^{-5} in this case. These tuning curves depend on the gain curves, respectively. In the case of large-diameter pumping and seeding beams, the tuning curve has been shifted toward lower frequency (longer wavelength); the output peak power in the sub-terahertz range has been increased. These are because the parametric gain in the low-frequency (long-wavelength) region was increased by expanding the beam diameters as shown in **Figure 4**; that is, wavelength conversion in the region was effectively achieved. Meanwhile, in the high-frequency (short-wavelength) region, both tuning range and output peak power have decreased, because of the large absorption associated with increasing propagation distance in the crystal.

Figure 7. A beam profile of a terahertz wave from the is-TPG measured by a terahertz-wave imager (IRV-T0831, NEC). The terahertz wave is focused by the $f = 50$ mm lens. The spot size is less than 220 mm (FWHM) at 1.5 THz.

Figure 8. An example of the wavelength and linewidth measurements. In this case, the wavelength (frequency) of the terahertz wave is approximately 398 μm (0.754 THz), and the linewidth is less than 5 GHz. This linewidth is near the Fourier transform limit in the sub-nanosecond pulse.

Figure 7 shows a beam profile of terahertz wave from the is-TPG measured by an imager for terahertz wave. The generated terahertz wave is collimated and focused by the Tsurupica cylindrical ($f = 100$ mm) and aspherical ($f = 50$ mm) lens. When the wavelength of the terahertz wave is $200\text{ }\mu\text{m}$, the spot size is less than $220\text{ }\mu\text{m}$ (full width at half maximum). We estimated the M^2 value less than 1.1, and the brightness was $B = Pp/(\lambda M^2)^2 \sim 0.2\text{ GW/sr}\cdot\text{cm}^2$. The intensity and electric field were 0.3 GW/cm^2 and 0.5 MV/cm at around 2.0 THz , respectively, at the focused point.

Figure 8 presents an example of wavelength and linewidth measurement by a scanning Fabry-Perot etalon consisting of two metal mesh plates. The horizontal axis represents the distance between metal meshes, and the vertical axis represents the energy of the transmitted terahertz wave. The metal meshes were made of nickel and had periods of $45\text{ }\mu\text{m}$ and reflectance of about 98% at 0.75 THz . As the distance between metal mesh plates increases, intensity peaks are observed periodically. In this case, the estimated wavelength (frequency) of the terahertz wave is about $398\text{ }\mu\text{m}$ (0.754 THz), and the linewidth is less than 5 GHz . This linewidth is a near the Fourier transform limit for the terahertz-wave pulse with a sub-nanosecond duration.

5. Conclusion

We have introduced here high-peak-power, narrow-linewidth, and continuously tunable terahertz-wave generation via wavelength conversion in a $\text{MgO}:\text{LiNbO}_3$ crystal. These result from the suppression of the SBS in a nonlinear crystal by using sub-nanoseconds pumping pulse. The high-brightness and continuously tunable source is important for the power calibration of terahertz-wave detectors. In general, the power calibration is based on calorimetry as a traceable international standard, but there is no power standard in the terahertz region. In this case, the power levels obtained from two kinds of pre-calibrated detectors, a calorimetric device, and a pyroelectric device, using the same terahertz beam were comparable [30]. Surprisingly, this was easily perceived directly by touch; the terahertz wave was felt to be similar to a 100 Hz (rep. rate) stimulation. Under our experimental conditions, the observed conversion efficiency is about 10^{-4} because the terahertz wave generated inside the crystal is absorbed by the nonlinear crystal itself while propagating to the crystal surface and is affected by Fresnel loss on the boundary surfaces. Furthermore, the parametric gain (absorption) of the terahertz wave in LiNbO_3 could be increased (decreased) by cooling the crystal [31]. The conversion efficiency improves by a factor of at least 10 at liquid nitrogen temperatures. In this case, under the condition of a pumping energy of 50 mJ/pulse , the expected brightness, brightness temperature, peak power, and electric field of the terahertz wave are greater than $4\text{ GW/sr}\cdot\text{cm}^2$, 10^{19} K , 1 MW , and 2 MV/cm , respectively, from our narrowband and continuously tunable is-TPG. Some applications require high-brightness terahertz waves, such as observing two- or multiphoton absorption to specific excitation states [32, 33]. The generation of the extremely high-brightness (megawatts ($\sim\text{MW}$) peak power and narrow ($\sim\text{GHz}$) linewidth) quasi-monochromatic terahertz-wave (several hundreds of cycles) pulses with field levels in the megavolt per centimeter ($\sim\text{MV/cm}$) range will enable novel applications in the field of terahertz nonlinear optics. We also introduced how to optimize the tuning curve of the is-TPG by controlling the pumping intensity and the interaction volume.

In the future, we have to endeavor to generate higher-brightness beam and wider tuning range for applied researches. Since extreme high-brightness terahertz-wave generation has attracted attention in recent years as a method of enabling nonthermal free target energy-level control and measurement. When we realize such a terahertz-wave control and measurement system, new applications in the terahertz region would be possible, and various issues in modern society could potentially be overcome. This system could be powerful tools not only for solving real-world problems but also fundamental physics, such as remote sensing, real-time spectroscopic measurement/imaging, 3D fabrication, and manipulation or alteration of atoms, molecules, chemical materials, proteins, cells, chemical reactions, and biological processes. We expect that these methods will open up new fields and unique applications.

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