TOPICAL REVIEW

Terahertz wave parametric source

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Abstract
Widely tunable terahertz (THz) wave generation by optical parametric processes based on laser light scattering from the polariton mode of nonlinear crystals is reviewed. Using parametric oscillation of LiNbO₃ or MgO-doped LiNbO₃ crystal pumped by a nanosecond Q-switched Nd: YAG laser, we have realized widely tunable coherent THz-wave sources in the range between 0.7 and 3 THz, with a simple configuration. For the efficient coupling of the THz wave, a monolithic grating coupler or an Si-prism array coupler was used. We report the detailed characteristics of the oscillation and the radiation, including tunability, spatial and temporal coherency, uni-directivity, and efficiency. Further, Fourier transform limited THz-wave spectrum narrowing was achieved by introducing the injection seeding method. A linewidth of about 100 MHz (0.003 cm⁻¹) was assured by the absorption spectrum measurement of low-pressure water vapour. At the same time, the THz-wave output was increased hundreds of times higher than that of a conventional generator which has no injection seeder. In addition, a wider tunability was observed using a tunable diode laser as the injection seeder. This room-temperature-operated, tabletop system promises to be a new widely tunable THz-wave source that is suited to a variety of applications.

1. Introduction

In recent years, the generation of terahertz (THz) radiation by optical rectification or photoconductive switching has been extensively studied using femtosecond laser pulses [1, 2]. Applied research, such as time domain spectroscopy, makes use of the high time resolution of THz waves and ultra broad bandwidth up to the THz region. In contrast, our research focuses on the development of tunable THz-wave sources with high temporal and spatial coherence using the resonant frequency of ferroelectric crystal lattices. Specifically, widely tunable coherent sources have a wide range of applications, such as in material science, solid state physics, molecular analysis, atmospheric research, bioscience, chemistry, gas tracing, material testing, food inspection, differential imaging, etc. Novel tunable sources already exist in the sub-THz (several hundred GHz) frequency region, such as the backward-wave oscillator (BWO). However, a widely tunable THz-wave source has long been desired in the frequency region above 1 THz, where the tuning capability of a BWO rapidly decreases.

Widely tunable sources covering the region between 1 and 3 THz (with wavelengths from 100 to 300 µm) are limited to free electron lasers or photomixers. The University of California at Santa Barbara (UCSB) plays a central role in exploring the THz region with free electron lasers [3]. However, the number of researchers who can access such an apparatus is limited, due to its large scale. In photomixers, two optical lasers are mixed to generate tunable THz waves.
This should become a reliable source if the limitation on the output due to the nondestructive limit of a photomixer is overcome. A p-type germanium (p-Ge) laser is another widely tunable source in this region, but it is primarily for use in pure science since liquid He is required [6, 7]. Although the aforementioned widely tunable sources can be used at the laboratory level, they do not satisfy all of the needs of researchers interested in practical applications. Therefore, there is significant potential for an explosion of applied research on the THz band if simple, convenient tunable sources are made available.

Many research efforts have been carried out a couple of decades ago concerning the generation of tunable coherent far-infrared radiation based on optical technology [8, 9]. Among them, THz oscillation and amplification were expected by decades ago concerning the generation of tunable coherent far-infrared waves. LiNbO₃, LiTaO₃, and GaP, which are both infrared-active and Raman-active, are made available.

The principle of tunable THz-wave generation is as follows. Polaritons exhibit phonon-like behaviour in the resonant frequency region (near the TO-phonon frequency ω_TO). However, they behave like photons in the nonresonant low-frequency region (figure 1), where a signal photon at THz frequency (ω_S) and a near-infrared idler photon (ω_I) are created parametrically from a near-infrared pump photon (ω_P). The pump photon is generated by the parametric process due to the nonlinearity of the material. The tuning is accomplished by controlling the angle of incidence of the pump photon and the vibrational mode. At the same time, the THz wave (signal) is generated by the parametric process due to the nonlinearity arising from both electronic and vibrational contributions of the material. The tuning is accomplished by controlling the propagation direction. Although the interaction is highly efficient, it should be noted that most of the generated THz waves are absorbed or totally reflected inside the crystal due to a large absorption coefficient, as well as a large refractive index in the THz range. To allow the THz radiation, a cut exit window is made in the corner of the crystal.

It is to our surprise that no research has been reported on this novel method since 1976 due to the alternative and successful use of submillimetre molecular gas lasers. In these six years, we have developed an efficient and widely tunable source of coherent THz waves based on the principle of previous works, and far better characteristics by introducing the monolithic grating coupler, the arrayed Si-prism coupler, doped crystals, and injection seeding.

2. Principles of operation

2.1. THz-wave parametric generation using a polariton

The generation of coherent tunable THz waves results from the efficient parametric scattering of laser light via a polariton (stimulated polariton scattering). A polariton is a quantum of the coupled phonon–photon transverse wave field, and stimulated polariton scattering occurs when the pump excitation is sufficiently strong in polar crystals, such as LiNbO₃, LiTaO₃, and GaP, which are both infrared-active and Raman-active. The scattering process involves both second- and third-order nonlinear processes. Thus, strong interaction occurs among the pump, the idler, and the polariton (THz) waves. LiNbO₃ is one of the most suitable materials for generating THz waves efficiently because of its large nonlinear coefficient (d33 = 25.2 pm V⁻¹, λ = 1.064 μm [15]) and its transparency over a wide wavelength range (0.4–5.5 μm). LiNbO₃ has four infrared- and Raman-active transverse optical (TO) phonon modes, called A₁-symmetry modes, and the lowest mode (ω_TO ~ 250 cm⁻¹) is useful for efficient tunable far-infrared generation because it has the largest parametric gain as well as the smallest absorption coefficient [16].

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\[ \omega_P = \omega_S + \omega_I \] (pump; T: THz; i: idler).

In the stimulated scattering process, the momentum conservation law \( \mathbf{k}_p = \mathbf{k}_i + \mathbf{k}_T \) (noncollinear phase-matching condition; see the insets of figure 1) also holds. This leads to the angle-dispersive characteristics of the idler and THz waves. Thus, a coherent THz wave is generated efficiently by using an optical resonator for the idler wave, and continuous and wide tunability is accomplished simply by changing the angle between the incident pump beam and the resonator axis.

2.2. Theory of THz-wave parametric gain

In stimulated polariton scattering, four fields mutually interact: the pump \( E_p \), idler \( E_i \), THz wave \( E_T \), and ionic vibration \( Q_0 \) (lowest A₁ mode). The parametric gain coefficients for the idler and THz waves are obtained by solving the classical coupled-wave equations that describe this phenomenon. Assuming a steady state and no pump depletion, the coupled-wave equations are written as [16–18]

\[
\begin{align*}
\nabla^2 + \frac{\omega^2}{c^2} \varepsilon_T E_T &= -\frac{\omega_0^2}{c^2} \chi_P E_p E_i^* \\
\nabla^2 + \frac{\omega_i^2}{c^2} \left( \varepsilon_i + \chi_R |E_p|^2 \right) E_i &= -\frac{\omega_0^2}{c^2} \chi_P E_p E_T^*
\end{align*}
\]

where \( \omega_0 (=\omega_P - \omega) \) and \( \omega \) denote the frequencies of the idler and THz wave, respectively, \( \varepsilon_p (p = T, i) \) is the permittivity in the material (LiNbO₃), and \( c \) is the velocity of light in vacuum.
The nonlinear susceptibilities $\chi_P$ and $\chi_R$ denote parametric and Raman processes, respectively, and are expressed as

$$\chi_P = d_E + \frac{S_0 \alpha_0^2}{\omega_0 - \omega^2} \cdot d_Q$$

(2)

$$\chi_R = \frac{S_0 \alpha_0^2}{\omega_0^2 - \omega^2 - i \alpha_0 \Gamma_0} \cdot d_Q^2$$

(3)

where $\alpha_0$, $S_0$, and $\Gamma_0$ are the eigenfrequency, oscillator strength, and damping coefficient (or linewidth) of the lowest $A_1$-symmetry phonon mode, respectively. The coefficients $d_E (=16\pi d_{33})$ and $d_Q$ denote the second- and third-order nonlinear processes, which originate from electronic and ionic polarization, respectively. According to the rate equation analysis, the expression of $d_Q$ in cgs units is given by

$$d_Q = \left[ \frac{8\pi c^4 n_p (S_{33}/L \Delta \Omega_0)}{S_0 \hbar \omega_0 \alpha_0^2 n_i (n_i + 1)} \right]^{1/2}$$

(4)

where $n_p$ ($\beta = p, i$) is the refractive index and $n_i = (exp[\hbar \omega_0/kT] - 1)^{-1}$ (h: Planck constant; k: Boltzmann constant; T: temperature) is the Bose distribution function.

The quantity $(S_{33}/L \Delta \Omega_0)$ denotes the spontaneous Raman (Stokes) scattering efficiency of the lowest $A_1$-symmetry phonon mode, where $S_3$ is the fraction of incident power that is scattered into a solid angle $\Delta \Omega$ near a normal to the optical path length $L$, and is proportional to the scattering cross section.

The coupled-wave equations (1) can be solved using the plane wave approach, and analytical expressions of the exponential gain for the THz wave and idler are

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

(5)

where $\phi$ is the phase-matching angle between the pump and the THz wave, $g_0$ is the low-loss limit, and $\alpha_T$ is an absorption coefficient in the THz region. In cgs units, they are written as

$$g_0 = \left( \frac{\pi \omega_0 I_p}{2c^3 \hbar n_i n_p} \right)^{1/2} \chi_P$$

(6)

$$\alpha_T = 2 |\text{Im} k_T| = \frac{2\omega}{c} \text{Im} \left( \varepsilon_{\infty} + \frac{S_0 \alpha_0^2}{\alpha_0^2 - \omega^2 - i \alpha_0 \Gamma_0} \right)^{1/2}$$

(7)

The low-loss parametric gain $g_0$ has the same form as the parametric gain in the optical region [19], but the nonlinear susceptibility $\chi_P$, which involves both second- and third-order processes (equation (2)), is almost entirely determined by the third-order (ionic) $d_Q$ term (more than 95% contribution).

The physical meaning of the susceptibility $\chi_P$ is explained as follows. According to the simple classical picture, a polar crystal, such as LiNbO$_3$, is considered to be an ensemble of individual molecular systems, where each molecule consists of nuclei bonded together and surrounded by an electron cloud. When the crystal is irradiated by the pump laser polarized along the $z$-axis of the crystal, the relatively light electron cloud absorbs the pump energy and follows the incident field, and the electronic dipole moment appears due to the displacement of the electron charge cloud with respect to the nuclei. This is the origin of the second-order nonlinearity expressed by the $d_E$ (or $d_{33}$) coefficient. Next, some of the energy absorbed by

the electrons is transferred to the nuclei, and two nuclei begin to vibrate (i.e. phonons are created) because the $A_1$-symmetry modes are infrared-active. This phenomenon leads to the ionic dipole moment or the third-order nonlinearity expressed by the Raman susceptibility $\chi_R$ (Stokes process). Finally, the ionic vibration modulates the electronic vibration, and an electron–ion interaction occurs because the vibrations are along the $z$-axis. Thus, the three-photon parametric scattering process involving the pump, Stokes (idler), and THz (polariton) waves involves both second- and third-order nonlinearities.

Figure 2 shows the calculated parametric gain $g_T$ for LiNbO$_3$ at typical pump intensities. A gain in the order of several cm$^{-1}$ is feasible in the frequency domain up to 3 THz at room temperature, and the gain is enhanced by cooling the crystal [20]. The decrease in the linewidth $\Gamma_0$ of the lowest $A_1$-symmetry phonon mode [21] makes the major contribution to the enhancement at low temperature, because $\alpha_T$ is nearly in proportion to $\Gamma_0$ (equation (7)). The reduced linewidth reduces the absorption coefficient $\alpha_T$ at THz frequencies, enhancing the parametric gain $g_T$, which is a monotonically decreasing function of the absorption coefficient (equation (5)).

Physically, when the polariton damping caused by random thermal activation is reduced and the polariton has a longer lifetime, an efficient parametric interaction occurs. It is also possible to increase the parametric gain, either by increasing the pump intensity or by using a shorter-wavelength pump source, because the gain is a monotonically increasing function of $g_0$, which is proportional to $[\omega_p - \omega_T \cdot I_p]^{1/2}$.

3. THz-wave parametric oscillator (TPO) with monolithic grating coupler

Widely tunable THz-wave generation was successfully demonstrated utilizing a grating coupler fabricated on the surface of an LiNbO$_3$ crystal which was pumped by a Q-switched Nd : YAG laser. In this section, we describe the detailed characteristics of the oscillation and the radiation including tunability, spatial and temporal coherency, and directivity.
3.1. TPO

Tunable THz-wave generation, using nonlinear optical methods, has been widely reported [8, 9] where difference-frequency mixing between two laser sources was utilized, though the conversion efficiency observed was poor. In contrast, higher conversion efficiency was obtained by simultaneous Raman and parametric oscillation, utilizing the polariton mode scattering of LiNbO$_3$ based on the $248\text{ cm}^{-1}$ $A_1$-symmetry soft mode [12–14]. This method used a single fixed optical source, and was performed at room temperature. The idler (Stokes) and signal (THz) waves were generated from the pump (near-infrared) wave in the direction consistent with the noncollinear phase-matching condition inside the LiNbO$_3$ crystal. The idler wavelength was longer than the pump wavelength by a few nanometres. A widely tunable coherent THz wave could easily be generated by slightly changing the incident angle of the pump beam. The nonlinearity arises from both the electronic and vibrational contributions of the material. Although the interaction between waves was generated by stimulated oscillation, most of the generated THz wave was absorbed or totally reflected inside the crystal due to the material’s large absorption coefficient and large refractive indices (5.2 at the THz range [22]). Therefore, it was rather difficult to couple out the THz radiation efficiently to the free space. In previously reported work [14], a specially prepared crystal was used, one corner of which was cut and polished at the proper angle to allow the THz radiation to emerge approximately normal to the exit surface, so that the problem of total internal reflection was avoided. In the work reported here, a grating structure on the surface of LiNbO$_3$ was employed to couple out the THz wave directly to the free air space with almost thousand times higher efficiency [23, 24].

3.2. Experimental methods

Our experimental setup is shown in figure 3. A 3.5 mm thick LiNbO$_3$ z-plane was cut to a dimension of $50(x) \times 10(y) \times 3.5(z) \text{mm}^3$. Two end surfaces in the x-plane were cut parallel, polished, and antireflection (AR)-coated for operation at $1.07\mu\text{m}$. The grating coupler was fabricated on the y-surface by the precise machining using a DISCO cutter (DAC-2SP/86) as shown in figure 4, and the grating pitch and length were 125 $\mu$m and 10 mm, respectively. Four different depths of grating, 20, 40, 60, and 100 $\mu$m, were formed on the sample to investigate the coupling efficiency. The crystal was placed inside the 15 cm long cavity as shown in figure 3, which was resonated by the idler wave using two high-reflection mirrors, M1 ($f = \infty$) and M2 ($f = 10\text{ m}$). Both mirrors were half-area-coated, so that only the idler wave could resonate and the pump beam propagate through the uncoated area without scattering. The mirrors and crystal were mounted on a rotating stage, and tunability was obtained by rotating the stage slightly to vary the angle of the resonator with respect to the pump wave. The pump source was a Q-switched Nd: YAG laser (SOLAR LF113, 1.064 $\mu\text{m}$) whose electric field was along the z-axis of the crystal. The pump power, pulsewidth, and repetition rate were 30 mJ/pulse, 25 ns, and 16.7 Hz, respectively. The pump beam entered the x-surface of the crystal and passed through the LiNbO$_3$ crystal close to the surface of the grating coupler to minimize the absorption loss of the THz radiation ($\alpha > 10\text{ cm}^{-1}$). A near-infrared idler oscillation around $1.07\mu\text{m}$ was clearly recognized by its oscillating spot above a threshold pump power density of about $130\text{ MW cm}^{-2}$. The THz-wave radiation was monitored with a 4 K Si bolometer (Infrared Laboratories). In order to suppress its response at infrared regions, a Yoshinaga filter was installed inside. A white polyethylene lens ($f = 60\text{ mm}$) was set in front of the bolometer to focus the THz-wave radiation. A Schottky barrier diode [25] was also utilized to detect the

![Figure 3. Experimental cavity arrangement for the TOP using a monolithic grating coupler on the y-surface of the LiNbO$_3$ crystal.](image3.png)

![Figure 4. Magnified view of the grating coupler formed on the y-surface of the LiNbO$_3$ crystal.](image4.png)
temporal characteristic of THz waves whose time response was faster than 10 GHz. At the same time, the intensity of the idler was measured by a power meter and recorded together with the bolometer output in a computer via a digital oscilloscope.

As shown in the inset of figure 3, the THz wave was generated in the direction satisfying the noncollinear phase-matching condition. Here, \( k_j \) is the wave vector with \( j = p, i, \) and \( T \), indicating the pump, idler, and THz waves, respectively. As the relationship \( k_p > k_i \gg k_T \) holds, the angle \( \phi \) between the pump and idler waves is small (\( \phi \approx 1^\circ \)), while the angle \( \delta \) between the idler and THz waves is large (\( \delta \approx 65^\circ \)).

### 3.3. Oscillation Characteristics

By varying the incident angle of the pump beam from 2.1° to 0.9°, the angle \( \phi \) between the pump and idler inside the crystal was changed from approximately 1° to 0.5°. As the phase-matching angle changed, the idler and the THz wavelengths were tuned from 1.072 to 1.068 \( \mu \)m and 140 to 310 \( \mu \)m, respectively, as depicted in figure 5. The angle \( \delta \) between the THz and idler inside the crystal changed from 66° to 65°. We also found that LiTaO₃ was capable of oscillating in the same cavity. In the case of LiTaO₃, we needed to make the incident angle of the pump larger than that of LiNbO₃.

For the 25 ns pulsewidth of the pump wave, the pulsewidths of the idler and THz waves were about 10 ns. Hence, the THz wave consists of several tens of thousands of cycles during the oscillation time, so that the coherency was sufficient. The THz wavelength and its linewidth were measured by a scanning Fabry–Perot etalon consisting of two Ni metal mesh plates with 65 \( \mu \)m grid. Figure 6 shows an example of the measurement. The displacement of one of the metal mesh plates corresponds directly to half of the wavelength. The free spectral range (FSR) of the etalon was about 100 GHz, and the linewidth was measured to be almost 20 GHz. The polarization of the THz wave was measured to be parallel to the \( z \)-axis of the crystal using a wire grid polarizer, as shown in figure 7, coinciding with those of the pump and idler waves. The THz-wave output from a 10 mm long grating coupler was measured to be 3 mW at peak with a pump power of 34.5 mJ/pulse. Accordingly, we could obtain almost 15 mW of output from the grating coupler on the whole \( y \)-surface of the crystal (50 mm length). We also made a cut exit at the end corner of the crystal; then we only got 5 \( \mu \)W at best, under the same conditions. In comparison to the cut exit, the grating coupler had an efficiency of better than \( \times 1000 \). Further, almost a hundred times higher THz-wave output can be obtained by cooling the crystal to liquid nitrogen temperature [20].

### 3.4. Radiation Characteristics of the Grating Coupler

Figure 8 shows the variation of the diffraction angle \( \theta \) (of the THz wave to the grating plane) with wavelength. The relation between the grating period \( \Lambda \) and the \( N \)th-order radiation angle \( \theta_N \) to the grating surface is given by \( \theta_N = \cos^{-1}(nT \cos \delta N\lambda T / \Lambda) \), where \( \delta \) is the incident angle to the grating and \( nT \) is the refractive index of LiNbO₃ at THz. The generated wavelengths ranged between 180 and 270 \( \mu \)m, while the radiated angles varied from 45° to 80°. This was
in good agreement with our prior calculation. The intensity distribution of the generated beam was Gaussian-like both along and perpendicular to the grating, as can be seen in figure 9. FWHM spot sizes of 8 and 12 mm were observed along the grating direction (x) at the measured points of 150 and 300 mm from the grating, which corresponds to a beam divergence of 0.8°. Along the z-axis, the divergence was determined by the pump spot size, and it was 1.5°. These results are in good agreement with diffraction theory.

The THz-wave absorption coefficient of LiNbO₃ at certain wavelengths was also measured. Keeping the angle condition between the pump and the idler constant, the THz-wave intensity was measured as a function of crystal displacement d along the y-axis. Figure 10 indicates the relation between the distance d and the intensity of the THz wave radiated from the grating coupler. Then the absorption coefficient was given by the slope to be \( \alpha_e = 21.2 \, \text{cm}^{-1} \) for \( \lambda_{\text{THz}} = 213 \, \mu\text{m} \), and \( \alpha_e = 51.9 \, \text{cm}^{-1} \) for \( \lambda_{\text{THz}} = 184 \, \mu\text{m} \) in another measurement; both were in good agreement with the previously reported values [22].

4. Arrayed Si-prism coupler for a TPO

In generating a THz wave by parametric oscillation in an LiNbO₃ crystal, some way of extracting the THz wave from the crystal is necessary, since the refractive index of the LiNbO₃ crystal for the THz wave is large enough to cause total internal reflection. We introduced an Si-prism coupler (n ≈ 3.4) to extract the THz wave generated inside a nonlinear crystal, thereby substantially improving the exit characteristics [26]. This section describes the characteristics of the oscillation, and a novel coupling method for THz waves using an arrayed Si prism [27]. Using the arrayed-prism coupler, there is a six-fold increase in coupling efficiency and a 40% decrease in the far-field beam diameter, compared with using a single-prism coupler. We also discuss the negative effect of the free carriers at the Si-prism surface excited by the scattered pump beam, and the positive effect of cavity rotation on the uni-directivity of THz-wave radiation from the Si prism.

4.1. Experimental methods

The basic configuration of the source consisted of a Q-switched Nd: YAG laser (LOTIS LS-2136, 1.064 \( \mu \text{m} \)) and a parametric oscillator, as shown in figure 11. The LiNbO₃ crystal used in the experiment was cut from a wafer \( 5 \times 65 \times 6 \, \text{mm}^3 \). The x-surfaces at both ends were mirror-polished and antireflection-coated. The y-surface was also mirror-polished, in order to minimize the coupling gap between the prism base and the crystal surface, and to prevent scattering of the pump beam, which excites a free carrier at the Si-prism base. The pump wave passed through the crystal close to the y-surface, to minimize the travel distance of the THz wave inside the crystal. The idler wave was amplified in an oscillator consisting of flat mirrors with a half-area HR coating. In the experiment, the beam diameter, pulsewidth, and repetition of the pump wave were 1.5 mm, 25 ns, and 50 Hz, respectively, and the typical excitation intensity was 30 mJ/pulse. The mirrors and crystal were installed on a precise, computer-controlled rotating stage (Nanoradian Stage, Harmonic Drive Systems Inc.) for the precise tuning.
A Si-prism array was introduced to obtain higher THz-wave output by increasing the coupling area. An array of seven Si-prism couplers was placed on the y-surface of LiNbO3, as shown in figure 11. The right-angle prisms were fabricated from high-resistivity Si ($\rho > 1 \, \text{k} \Omega \, \text{cm}$, $\alpha \cong 0.6 \, \text{cm}^{-1}$); each was cut from a bulk Si crystal, using a precise diamond cutter, to dimensions of $8.0 \times 6.1 \times 5.1$ (face) $\times 5.0$ (thickness) mm, and the angles were $50^\circ$–$40^\circ$–$90^\circ$. The total base length was $8 \, \text{mm} \times 7 = 56 \, \text{mm}$. A prism opening angle of $\xi = 40^\circ$ was chosen so that the THz wave would emerge almost normal to the prism face ($6.1 \times 5.0 \, \text{mm}^2$). The base of the prisms was pressed by a specially designed holder against the y-surface of the LiNbO3 crystal to maximize coupling efficiency.

4.2. Experimental results

Typical input–output characteristics of a TPO with an Si-prism array are shown in figure 12, in which the oscillation threshold was $18 \, \text{mJ/pulse}$. With a pump power of $34 \, \text{mJ/pulse}$, the THz-wave output from the prism array was $192 \, \text{pJ/pulse}$ ($\cong 19.2 \, \text{mW}$ at the peak), calibrated using the sensitivity of the bolometer. Since the Si-bolometer output becomes saturated at approximately $5 \, \text{pJ/pulse}$, we used several sheets of thick paper as an attenuator after calibration. The minimum sensitivity of the Si bolometer is approximately $1 \, \text{fJ/pulse}$; therefore, the dynamic range of measurement using the TPO as a source is $192 \, \text{pJ/1 fJ}$, which exceeds $50 \, \text{dB}$. In the case of single-prism coupling, the typical output was about $30 \, \text{pJ/pulse}$ ($3 \, \text{mW}$ at the peak) at best under similar conditions: THz wavelength $180 \, \mu\text{m}$ and idler output $1 \, \text{mJ/pulse}$. In comparison, the prism array was capable of emitting more than six times as much THz-wave energy as the single-prism coupling due to seven times wider base area.

A small portion of the pump beam was reflected from the end mirror or scattered at the crystal edge and shone on the emitting face of the Si prism, generating a free carrier that strongly absorbed the THz wave. An HR mirror was therefore installed in front of the last Si prism (rightmost in figure 11) to intercept the reflected pump beam, as illustrated in figure 11. The HR mirror needed to be close to the y-surface of the LiNbO3 crystal to intercept the pump beam, as only $10 \, \mu\text{J/cm}^2$ of pump is enough to generate the free carrier in Si [28]. Without the HR mirror, the THz-wave output from the last Si prism decreased to $10^{-2}$–$10^{-3}$ of the output with the HR mirror in place. Because of the difficulty in perfectly shielding the scattered pump, it was difficult to obtain maximum THz-wave output ($\cong 3 \, \text{mW}$) using a single-prism coupling, even with the HR mirror. On the other hand, it is much easier to extract maximum THz-wave output ($\cong 20 \, \text{mW}$) from the arrayed prism, because the last prism acts as a perfect auxiliary shield for the scattered pump.

The spatial intensity distribution of the THz-wave radiation was measured by transversely shifting a Si bolometer with a 1.4 mm wide incident slit. The beam pattern and diffraction of the THz wave in the z-plane are shown in figures 13 and 14 for the single- and arrayed-prism couplers, respectively. In both figures, $d$ indicates the distance between the prism coupler and the slit. Both measurements, the THz wavelength was $170 \, \mu\text{m}$ and the angle between the THz beam and the y-surface of the crystal was $50^\circ$.

With single-prism coupling, both the near- and far-field patterns were Gaussian-like, as shown in figure 13, and the diffraction angle in the far field was measured to be $1.4^\circ$. With arrayed-prism coupling, the far-field pattern was almost Gaussian-like, whereas the near-field pattern was asymmetric, as shown in figure 14. In the near field, higher output was observed from the prisms closest to the pump exit surface (right in figures 11 and 14), because as the distance between the pump and the y-surface of the crystal shortens, the absorption loss of the THz wave decreases. The output of each prism was distinguishable at $d < 10 \, \text{cm}$, whereas the beam pattern...
became continuous at \( d > 20 \text{ cm} \). The diffraction angle of the THz wave emitted from the prism array coupler was apparently smaller than that of single-prism coupling, comparing the FWHM of the beam pattern at distance \( d = 100 \text{ cm} \). At \( d = 100 \text{ cm} \), the FWHM is 58 mm for the single-prism coupling and 34 mm for the arrayed-prism coupling. The far-field diffraction angle is decided by the emitting aperture width and the wavelength. The smaller diffraction angle was obtained by the prism array due to the seven times wider emitting aperture than the single prism.

In previous experiments [14], a cut exit was used to avoid total internal reflection, as illustrated in figure 15. A cut exit was made at the corner of the LiNbO\(_3\) crystal, so that the THz wave emerged approximately normal to the exit surface. In this case, the refractive index dispersion of LiNbO\(_3\) and change of the phase-matching angle, \( \delta \), directly influenced the THz-wave direction change, \( \Delta \theta_c \). On the other hand, when an Si-prism coupler is used, radiation is almost in one direction and variation in the phase-matching angle is substantially reduced. Using the phase-matching conditions, the output angle of the THz wave \( \theta_p \) yields [29]

\[
\sin \theta_p = n_c^2 \sin \left[ \arcsin \left( \frac{n_i}{2n_T^2} + \frac{n_i \lambda_T}{2n_T^2(\lambda_T - \lambda_p)} \right) - \frac{n_T^2 \lambda_p}{2n_T^2 n_i (\lambda_T - \lambda_p)} - \frac{(n_i^2 - n_p^2) \lambda_T^2}{n_c^2 n_i (\lambda_T - \lambda_p)^2} - \xi \right]
\]

where \( \lambda_p \) and \( \lambda_T \) are the pump and THz wavelength, \( n_p \), \( n_i \), and \( n_T \) are the refractive indices of the pump, idler, and THz waves in LiNbO\(_3\), \( n_c^2 \) is the refractive index of the THz wave in silicon, and \( \xi \) denotes the opening angle of the Si prism. Figure 15 shows the calculated changes in radiation angle for these two methods of coupling. The changes in the radiation angle are set at zero for \( \lambda_{THz} = 200 \mu \text{m} \), for comparison. The dotted and broken lines indicate changes in the radiation angle for the cut exit, \( \Delta \theta_c \), and Si-prism coupler, \( \Delta \theta_p \), respectively. The solid line shows the change in THz-beam direction when observed from outside the TPO. It is important to note that since the TPO cavity can be angle-tuned by rotating the stage in the direction counter to \( \Delta \theta_c \), the actual angle change becomes much less than \( \Delta \theta_p \). For the tuning range of \( 100-420 \mu \text{m} \), \( \Delta \theta_c = 16.5^\circ \), \( \Delta \theta_p = 4.0^\circ \), and the actual change, \( \Delta \theta_p - (\text{cavity rotation}) = 1.5^\circ \).

An example of the tuning range using the Si prism is shown in figure 16 as the solid line. In the figure, stars indicate the
The THz-wave output can be increased by increasing the pump beam diameter in the z-axis direction, so that the THz wave is coupled out from a wider area of the y-surface of the crystal. In this case, the diffraction angle of the THz wave in the vertical direction decreases, since it is decided by the diameter of the pump beam.

5. Injection-seeded THz-wave parametric generator

Coherent tunable THz waves were successfully generated using an injection-seeded THz-wave parametric generator (IS-TPG) based on laser light scattering from the A₁-symmetry polariton mode of MgO:LiNbO₃ crystals. A THz-wave spectrum narrowing to the Fourier transform limit was achieved by injection seeding the idler wave (near-infrared Stokes). This resulted in a THz-wave output approximately 300 times higher than that of a conventional TPG, which has no injection seeder. In addition, a wide tunability from 0.7 to 2.4 THz was observed using a tunable diode laser as an injection seeder. A resolution of less than 100 MHz (0.003 cm⁻¹) was assured by the absorption spectrum measurement of low-pressure water vapour. This compact system operates at room temperature and promises to be a new, widely tunable THz-wave source.

5.1. IS-TPG

We have researched a TPO and a TPG using LiNbO₃ or MgO:LiNbO₃ crystals. The TPO has proved to be a useful coherent THz-wave source that operates at room temperature. It is continuously tunable in the 1–3 THz range in one operation and can emit peak powers of up to several tenths of a mW. The difference between a TPO and a TPG is that the former has an idler cavity while the latter does not. The THz-wave linewidth of a conventional TPG exceeds 500 GHz and the THz-wave output is much smaller than that from a TPO. Therefore, we previously concentrated our efforts on the development of a TPO system, although its linewidth was tens of GHz.

This section is primarily concerned with new work on TPGs, which appear to perform better than TPOs. The TPG spectrum was narrowed to the Fourier transform limit of the pulsewidth by introducing injection seeding to the idler [30, 31]. The purity of the THz-wave frequency was dramatically improved to $\Delta \nu / \nu < 10^{-4}$. Simultaneously, the output obtained was several hundred times higher than that of a conventional TPG. In addition, wide tunability and fine resolution were demonstrated using a tunable seeder. Even in the optical region, injection seeding to a nanosecond (ns) optical parametric generator (OPG) has not been reported until recently [32], due to the limit of parametric gain.

5.2. Experimental setup

Figure 17 shows the setup of our experimental IS-TPG. Arrangements were tested using one, two, and three nonlinear crystals, 65 mm in length. The maximum THz-wave output was obtained when two crystals (5 mol% MgO-doped LiNbO₃) were used in series. The TPG efficiency of an MgO:LiNbO₃ is several times higher [16] than that of a nondoped LiNbO₃. Both crystals were cut into $65 \times 5 \times 5 \text{mm} (x \times y \times z$-axis) pieces. The $x$-surfaces were polished and antireflection-coated. An array of seven Si-prism couplers was placed on the $y$-surface of the secondary MgO:LiNbO₃ crystal for efficient coupling of the THz wave as shown in figure 17. The pump used was a single longitudinal mode (SLM) Q-switched Nd:YAG laser (Spectron SL404T, wavelength: 1.064 µm; energy: $<50 \text{mJ/pulse}$; pulsewidth: 15 ns; beam profile: TEM₀⁰). The pump beam diameter was decreased to 0.8 mm using a telescope in order to increase the power density. The pump power density was $<530 \text{ MW cm}^{-2}$ at the crystal surface and could be varied with an attenuator. The pump beam was almost normal to the crystal surfaces as it entered the crystals and passed through the crystal close to the $y$-surface. A continuous-wave (CW) SLM Yb-fibre laser (wavelength: 1.070 µm; power: $<300 \text{ mW}$) or tunable diode laser (wavelength: 1.066–1.074 µm; power: 50 mW) was used as an injection seeder for the idler. Observation of the intense
idler beam easily confirmed the injection-seeded THz-wave generation. The polarizations of the pump, seed, idler, and THz waves were all parallel to the $z$-axis of the crystals. The THz-wave output and temporal waveform were measured with a 4 K Si bolometer and a Schottky barrier diode detector, respectively.

### 5.3. Power enhancement

Energy enhancement of the THz and idler waves by injection seeding is shown in figures 18(a) and (b), respectively. The THz and idler outputs are roughly proportional to each other. Comparison of the output from 0 and 200 mW seeding enabled us to determine that the THz-wave and idler energy increased by factors of nearly 300 and 500, respectively. The maximum conversion efficiency was achieved when the pump and seed beams almost fully overlapped at the incident surface of the first MgO : LiNbO$_3$ crystal, as shown in figure 17. This was confirmed by the fact that initial excitation is an essential feature of injection seeding. The maximum THz-wave output of 900 pJ/pulse (peak > 100 mW) was obtained with a pump of 45 mJ/pulse and a seed of 250 mW. In our previous studies, the maximum THz-wave output from a conventional TPG and a TPO was 3 and 190 pJ/pulse, respectively [27]. The Si bolometer became saturated at about 5 pJ/pulse, so we used several thick calibrated papers as an attenuator. As the minimum sensitivity of the Si bolometer was almost 1 fJ/pulse, the dynamic range of the injection-seeded TPG system was 900 pJ to 1 fJ $\sim$ 60 dB, which is sufficient for most applications. The dynamic range can be significantly increased using a lock-in amplifier.

Figure 19 shows the THz-wave (a) and idler (b) outputs as functions of the seed power. The outputs began to saturate with a seed power of almost 100 mW. A relatively high seed power was required in this experiment because the seed energy did not fully contribute to the idler generation. The seed and idler beams were spatially separated from each other as shown in figure 20. This is because the pump and seed beams were spatially separated inside the secondary MgO : LiNbO$_3$ crystal (see figure 17), and because most of the idler energy was generated inside the secondary MgO : LiNbO$_3$ crystal.

![Figure 19. THz wave (a) and idler (b) outputs as a function of seed power.](image)

![Figure 20. Beam profiles of the seed and idler at a distance of 160 cm from the crystal end. They are separated from each other because most of the idler output is generated inside the secondary MgO : LiNbO$_3$ crystal (see figure 17).](image)
On the other hand, when one crystal was used, 10 mW of seed power was enough to obtain idler saturation because the pump and seed beams were not separated. Therefore, it is important to somehow confine the pump and seed beams in a long interaction volume, in order to decrease the required seed power and to increase the efficiency. In figure 20, the idler beam pattern was expanded in the z-axis direction probably due to the photorefractive effect inside the crystal. The angle between the idler beam and the crystal surface normal was almost 1.5°, proving that the cavity effect of the crystal surfaces has no relation to this parametric generation.

Figure 21 shows examples of temporal waveforms of the pump, idler, and THz wave using a pump energy of 45 mJ and a seed power of 250 mW. The pulsewidths of the pump and idler are 15 and 4 ns, respectively. The observed pump depletion (28.4%) was the largest depletion encountered during our TPG/TPO research. The THz waveform was also found to be depleted, probably due to back conversion of the pump. The second peak of the pump waveform in figure 21 is due to back conversion of the pump, and the product of $E_p$ and $E_i$ resulted in the second peak of the THz waveform. Depletion of the THz waveform was not observed with pump energies below 35 mJ/pulse. The THz waveform began to deplete as the pump energy increased, although the THz-wave energy continued to increase, as shown in figure 18, due to the pulsewidth expansion.

Figure 22 shows the THz-wave beam pattern in the horizontal (upper) and vertical (lower) directions, respectively, at a distance of ~40 cm from the Si-prism array. The beam pattern was nearly Gaussian and had a diameter of 7 mm, which is suitable for many applications. The original vertical divergence was about 6°, as determined from the pump beam diameter and the wavelength according to diffraction theory. A cylindrical lens ($f = 30$ mm) made of polymethylpentene (PMP or ‘TPX’) was used, as shown in figure 17, to collimate the THz-wave divergence in the vertical (z-axis) direction. As for the horizontal direction, the beam diameter decreased as it propagated, due to the phased array-like effect of the Si-prism array [27]. Furthermore, using a short focus TPX or Si lens, the THz beam can be tightly focused into spot of diameter ~0.5 mm.

5.4. Spectrum narrowing

Figure 23 shows the effect of injection seeding on idler spectrum narrowing. The dotted line indicates the idler spectrum of a conventional TPG without injection seeding, and the solid line indicates the idler spectrum of an injection-seeded TPG. The resolution limit of the spectrum analyser used was 0.2 nm, so the real idler spectrum was much narrower than that shown in this figure. Using a solid etalon, the idler spectrum was assured to be less than 1 GHz.

The THz wavelength and linewidth were measured using a scanning Fabry–Perot etalon consisting of two Ni metal meshes with a 65 μm grid. Figure 24 shows the transmitted THz-wave power as a function of etalon spacings of (a) ~80 mm and (b) ~210 mm. Figure 24(a) shows the spectrum and output during the 20 min scan. The displacement between the two periods (190 μm) directly corresponds to the wavelength. The merit of an injection-seeded TPG lies in its output stability due to the mode-hop-free characteristic, since it has no cavity. On the other hand, as with an injection-seeded TPO [33], the cavity length must be actively controlled to match the seed wavelength in order to stabilize the output. Since the etalon spacing was up to 210 mm, the THz-wave pulse (3.4 ns) made less than three round trips in the etalon cavity; thus, the resolution is inevitably limited.

The Fourier transform limit of the spectral width was calculated from the pulse shape of the THz wave as measured by SBD. The typical pulsewidth of the THz wave was 3.4 ns, as shown in figure 25(a), and was almost identical to that of the idler, which was measured with a high-speed photodetector. Figure 25(b) shows the power spectrum of the THz wave calculated from the upper graph, and indicates that the linewidth was 136 MHz. In this calculation, we ignored any fluctuations in the background noise near the zero level in figure 25(a). Figures 24 and 25 confirmed that the linewidth of the THz wave was narrowed to near the Fourier transform limit. The THz-wave linewidth was still more than 10 GHz,
Figure 23. Narrowing of the idler (1.07 µm) spectrum by injection seeding. The dotted and solid lines indicate the idler spectrum of a conventional TPG and an IS-TPG, respectively. The resolution limit of the spectrum analyser used was 0.2 nm, so the real idler linewidth was much narrower than in this figure.

Figure 24. THz linewidth and wavelength measured with a scanning Fabry–Perot etalon consisting of two metal mesh plates. (a) The stability of the spectrum is demonstrated and the displacement between the two periods (190 µm) corresponds directly to the wavelength. (b) The FSR of the etalon is 750 MHz and the linewidth of the THz wave is measured to be less than 200 MHz (0.0067 cm⁻¹), which is our measurement resolution limit.

Figure 25. (a) Temporal THz-wave output measured by the SBD, and (b) calculated Fourier transform limit of the spectral width from the measured temporal THz waveform. The typical pulsewidth of the THz wave was 3.4 ns (a), and the calculated linewidth was 136 MHz (b).

5.5. Wide tunability

It was possible to tune the THz wavelength using an external cavity laser diode as a tunable seeder. A wide tunability from 125 to 430 µm (frequency: 0.7–2.4 THz; wave number: 23–80 cm⁻¹) was observed as shown in figure 26 by changing both the seed wavelength and the seed incident angle. Open squares and closed circles indicate the tunability of the THz and idler waves, respectively. Both crystals were MgO : LiNbO₃ in this experiment. The wavelength of 430 µm (0.7 THz) was the longest ever observed during our study of TPGs and TPOs. In the longer-wavelength region, the angle between the pump and idler becomes less than 1°; thus it is difficult for the TPO to oscillate only the idler inside the cavity without scattering the pump. In the shorter-wavelength region, the THz output is comparatively smaller than the idler output, due to the larger absorption loss inside the crystal.

The absorption spectrum of low-pressure (<1 torr) water vapour was measured to demonstrate the continuous tunability and the high resolution of the IS-TPG. The absorption gas cell used was an 87 cm long stainless light pipe with TPX windows at both ends. Figure 27 shows an example of measurements at around 1.92 THz, where two neighbouring lines exist. A resolution of less than 100 MHz (0.003 cm⁻¹) was clearly shown. In fact, it is not easy for FTIR spectrometers in the THz-wave region to demonstrate a resolution better than 0.003 cm⁻¹ because of the instability of the scanning mirror for more than a meter. The system is capable of continuous tuning at high spectral resolution in 4 GHz segments anywhere even with an injection-seeded TPG that used a multi-frequency Nd : YAG laser as the pump. Thus, both the pump and seed must be SLM lasers to obtain the transform-limited THz wave with a TPG.
in the 0.7–2.4 THz region. The range of continuous tuning is currently restricted by the mode hop of the tunable laser diode. Since there is no cavity to be slaved, continuous tuning is extendible, in principle, to the full tunability of the IS-TPG by using a mode-hop-free seeder, such as a Littman-type external cavity diode laser.

Figure 28 shows the change in THz-wave output as a function of seed incident angle. The seed wavelength (1.070 µm) and generated THz wavelength (190 µm) were confirmed to be constant. The angle of incidence shows significant tolerance.

### 6. Conclusion

We reviewed the widely tunable THz-wave generation based upon the optical parametric process. We have demonstrated a TPO using monolithic grating couplers in section 3. Measurements on tunability, coherency, power, polarization, radiation angle, and divergence have been accomplished, proving this method to be suitable for many application fields. These include spectroscopy, communication, medical and biological applications, THz imaging, and so forth. In section 4, an efficient TPO was demonstrated by introducing an arrayed Si-prism coupler. The output was more than six times greater than with a single-prism coupling. The diffraction angle observed using the prism array was smaller than with single-prism coupling, because of the phased-array-like effect. Furthermore, we explained the uni-directional THz-wave radiation from the Si-prism coupler.

In section 5, we have demonstrated a high spectral resolution IS-TPG. We measured the power enhancement, spectrum narrowing, and tunability of this IS-TPG. In comparison with a conventional TPG without injection seeding, the output was increased from 3 to 900 pJ/pulse, and the linewidth was decreased from >500 GHz to ≈100 MHz. Wide tunability up to 430 µm (0.7 THz) was assured using a tunable seeder, and fine tuning was demonstrated by THz spectroscopy of low-pressure water vapour.

Further improvement of our system is possible. As OPGs and OPOs have improved tremendously in the last decade, the use of TPGs and TPOs shows great potential to move towards a lower threshold, higher efficiency, and wider tunability. A lower threshold and a narrower linewidth can be expected using a nonlinear optical waveguide and a longer pump pulsewidth, respectively. Operation in other wavelength regions, through proper crystal selection, should also be possible. Success in this will prove the practicality of a new widely tunable THz-wave source, the IS-TPG, that will compete with free-electron lasers and p-Ge lasers. For tunable THz-wave applications, the simplicity of the wave source is an essential requirement since cumbersome systems do not
encourage new experimental thoughts and ideas. Compared with the available sources, the present parametric method has significant advantages in compactness, tunability, and ease of handling.

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References

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