1 Terahertz-Wave Parametric Generator (TPG) / Pumping source

1.1 Injection-seeded THz-wave Parametric Generator

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ABSTRACT

Coherent tunable terahertz (THz) waves were successfully generated using an injection-seeded THz-wave parametric generator (TPG) based on laser light scattering from the A1-symmetry polariton mode of MgO:LiNbO3 crystals. 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. Resolution of less than 100 MHz (0.003 cm⁻¹) was assured by the absorption spectrum measurement of low-pressure water vapor. This compact system operates at room temperature and promises to be a new, widely tunable THz-wave source.

I. INTRODUCTION

The THz-wave (very far-infrared) region has attracted significant interest in recent years. The generation of THz radiation by optical rectification or photo-conductive switching has been extensively studied using femto-second laser pulses [1,2]. Applied research, such as time domain spectroscopy (TDS), 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. 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, etc. Novel tunable sources already exist in the sub-THz (several hundreds GHz) frequency region, such as a 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. Several candidate schemes have been reported [3-6], although they do not avoid one or more of the following problems: large size, operational difficulty, liquid He requirement, and low output power. A compact user-friendly source will inevitably find many applications in laboratory-based research, or even commercially.

Several decades ago, the author (Nishizawa) pointed out that far-infrared or THz-wave generation could be realized using the lattice vibrations in semiconductors [7]. We have researched a THz-wave parametric oscillator (TPO) and THz-wave parametric generator (TPG) based on the polariton mode scattering of LiNbO3 or MgO:LiNbO3 crystals [8-11]. The TPO has proved to be a useful coherent THz-wave source that operates at room temperature. It is continuously tunable in the 100- to 300-μm (1- to 3-THz) range in one operation and can emit peak powers of up to several tenths of a milli-Watt. 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 concentrated our efforts on the development of a TPO system, although its linewidth was several tens of GHz.

In this paper, the TPG spectrum was narrowed to the Fourier transform limit of the pulselength by introducing an injection seeding to the idler. 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. As far as we know, injection seeding to ns optical parametric generators (OPG) has not been reported until recently [12], due to the limit of parametric gain.

II. EXPERIMENTAL SETUP

Fig. 1 shows the setup of our experimental injection-seeded 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 (nondoped LiNbO$_3$ and 5 mol% MgO:LiNbO$_3$) were used in series. Although the TPG efficiency of MgO:LiNbO$_3$ is higher [13], its crystal quality is poorer than that of a nondoped LiNbO$_3$. Therefore, we used nondoped LiNbO$_3$ at the front as a seed amplifier. Both crystals were cut into $65 \times 6 \times 5$-mm ($x \times y \times z$-axis) pieces. The x-surfaces at both ends were polished, so that they were parallel, and coated with antireflection coating centered at 1.064 $\mu$m. The y-surface was also mirror polished to eliminate scattering of the pump beam and to minimize the coupling gap between the Si-prism base and the crystal surface. An array of seven Si-prism couplers was placed on the y-surface of the MgO:LiNbO$_3$ crystal for efficient coupling of the THz-wave [14]. The pump used was a single longitudinal mode (SLM) Q-switched Nd:YAG laser (wavelength: 1.064 $\mu$m, energy: $< 50$ mJ/pulse, pulsewidth: 15 ns, beam profile: TEM$_{00}$). 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 $< 530$ 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 MgO:LiNbO$_3$ close to the y-surface in order to minimize the absorption loss of the THz-wave inside the crystal ($\alpha > 10$ cm$^{-1}$). A continuous wave (CW) SLM Yb-fiber laser (wavelength: 1.070 $\mu$m fixed, power: $< 300$ mW) or tunable diode laser (wavelength: 1.066 - 1.074 $\mu$m, power: 50 mW) was used as an injection seeder for the idler.

![Fig. 1. The setup used for our experimental injection-seeded THz-wave parametric generator (TPG). The pump was a single longitudinal mode Q-switched Nd:YAG laser (1.064 $\mu$m), and the seed for the idler was a continuous-wave Yb-fiber laser (1.070 $\mu$m) or tunable laser diode (1.066 - 1.074 $\mu$m).](image)

As shown in the inset of Fig. 1, the idler and THz-waves were generated simultaneously in a direction that satisfied noncollinear phase-matching conditions. Here, $k_j$ is the wave vector with $j = p$, i, and T, which indicate the pump, idler, and THz waves, respectively. As the relationship $k_p > k_i > k_T$ holds, the angle between the pump and idler is small and the angle between the pump and THz-wave is large. For example, the angle between the pump and seed beams outside the crystal was almost 1.43°, in accordance with noncollinear phase-matching
conditions between the pump (1.064 \( \mu m \)), idler (1.070 \( \mu m \)) and THz-wave (190 \( \mu m \)). Idler observation with an IR visualizer easily confirmed the 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 4K Si-bolometer (Infrared Laboratories Inc.) and a Schottky barrier diode detector (SBD) [15], respectively.

III. POWER ENHANCEMENT

Energy enhancement of the THz and idler waves by injection seeding is shown in Figs. 2 (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 LiNbO\(_3\) crystal, as shown in Fig. 1. 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 [14], respectively. 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 – 60 dB, which is sufficient for most applications. The dynamic range can be significantly increased using a lock-in amplifier.

![Fig. 2. The input-output characteristics of a TPG, showing energy enhancement of the (a) THz-wave (190 \( \mu m \)) and (b) idler (1.070 \( \mu m \)) by a factor of several hundreds with injection seeding.](image)

Fig. 3 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 Fig. 4. This is because the pump and seed beams were spatially separated inside the MgO:LiNbO\(_3\) crystal (see Fig. 1), and because most of the idler energy was generated inside the MgO:LiNbO\(_3\) crystal. On the other hand, when one LiNbO\(_3\) 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 Fig. 4, the idler beam pattern was expanded in the z-axis direction probably due to the photo-refractive effect inside the crystal. The angle between the idler beam and the crystal x-surface normal was almost 1.5°, proving that the cavity effect of the crystal surfaces has no relation to this parametric generation.

![Fig. 4. The 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 MgO:LiNbO₃ crystal. (See Fig. 1)](image)

Fig. 5 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 [16]. 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 Fig. 5 is due to back conversion, and the product of $E_{\text{pump}}$ times $E_{\text{idler}}$ 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 Fig. 2, due to the pulsewidth expansion.

Fig. 6 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 mmΦ, 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 Fig. 1, 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 [14]. Furthermore, the THz beam can be tightly focused into a ~ 0.5 mmΦ spot using a short focus TPX or Si lens.
IV. SPECTRUM NARROWING

Fig. 7 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 analyzer used was 0.2 nm, so the real idler spectrum was much narrower than 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. Fig. 8 shows the transmitted THz-wave power as a function of etalon spacings of (a) ~ 80 mm and (b) ~ 210 mm [16]. Fig. 8 (a) demonstrates the stability of the spectrum and output during the 20-minute 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, the cavity-length must be actively controlled to match the seed wavelength in order to stabilize the output [17]. In Fig. 8 (b), the free spectral range (FSR) of the etalon is 750 MHz and the THz-wave linewidth was measured to be less than 200 MHz (0.0067 cm⁻¹), which is our measurement resolution limit. 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 Fig. 9 (a), and was almost identical to that of the idler, which was measured with a
Fig. 7. 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 injection-seeded TPG, respectively. The resolution limit of the spectrum analyzer used was 0.2 nm, so the real idler linewidth was much narrower than in this figure.

Fig. 8. The 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.

Fig. 9  (a) Temporal THz-wave output measured by the SBD, and (b) the 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, as shown in the upper figure, and the calculated linewidth was 136 MHz, as shown in the lower figure.
high-speed photo detector. Fig. 9 (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 Fig. 9 (a). Figs. 8 and 9 confirmed that the linewidth of the THz-wave was narrowed to near the Fourier transform limit. It is important to note that the THz-wave linewidth was still ~20 GHz, 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.

V. 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 to 2.4 THz, wave number: 23 to 80 cm\(^{-1}\)) was observed by changing both the seed wavelength and the seed incident angle as shown in Fig. 10. Squares and circles indicate the tunability of the THz and idler waves, respectively. Both crystals were MgO:LiNbO\(_3\) 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 range of continuous tuning is presently restricted to several GHz, due to the mode hop of the laser diode.

Fig. 11 shows an example of the absorption spectrum measurement of low-pressure (<1 torr) water vapor at around 1.919 THz. The gas cell used was an 87-cm-long stainless light pipe with TPX windows at both ends. Resolution of less than 100 MHz (0.003 cm\(^{-1}\)) was clearly shown. In fact, it is not easy for a typical FTIR spectrometers in the THz-wave region to show a resolution better than 0.003 cm\(^{-1}\) because of the instability of the scanning mirror.

Fig. 12 shows the change in THz-wave output as a function of the seed incident angle. In this experiment, the seed wavelength (1.07 μm) and THz wavelength (190 μm) were fixed, and the calculated noncollinear phase-matched angle was 1.43°. Here, it is important that injection seeding was not overly sensitive to the seed incident angle. In addition, the linewidth was assured to be less than 200 MHz at any deviated incident angle. From this, we see that continuous tuning is possible, to some extent, by simply varying the seed wavelength without having to adjust the incident angle. In practice, the tuning in Fig. 11 was produced without changing the seed incident angle. Tuning without mechanical movement will lead to a stable and compact spectroscopic system. Even when the incident angle must be varied for wide tuning such as in Fig. 10, there is no requirement to precisely control the angle due to this tolerance.

VI. CONCLUSION

In this paper, we have demonstrated a high spectral resolution, injection-seeded ns-TPG, based on the noncollinear phase matching in MgO:LiNbO\(_3\) and pumped at 1.064 μm. We measured the power enhancement, spectrum narrowing, and tunability of this 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 vapor.
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 narrower linewidth can be expected using a nonlinear optical waveguide and longer pump pulsewidth, respectively. Using a suitable seeder, wide range and mode-hop-free tuning is possible for a TPG, since there is no cavity and, hence, no requirement to actively control its length. 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 that will compete with free-electron lasers and p-Ge lasers.

![Graph 1](image1.png)

**Fig. 10.** The wide tunability of an injection-seeded TPG. Squares and circles indicate the tunability of the THz and idler waves, respectively.

![Graph 2](image2.png)

**Fig. 11.** An example of the absorption spectrum measurement of low-pressure (<1 torr) water vapor at around 1.919 THz. Resolution of less than 100 MHz (0.003 cm\(^{-1}\)) was clearly shown.

![Graph 3](image3.png)

**Fig. 12.** Variation in THz-wave output as a function of the seed incident angle. The seed wavelength (1.070 \(\mu m\)) and generated THz wavelength (190 \(\mu m\)) were confirmed constant. The angle of incidence shows significant tolerance.

**ACKNOWLEDGMENTS**
The authors thank Dr. K. Yamada of the National Institute for Resources and Environment, and Dr. H. Komine of TRW Space & Technology Division for useful discussions; Prof. K. Mizuno of the Research Institute of Electrical Communication, Tohoku University, for providing the Schottky barrier diodes; C. Takyu for his excellent work coating the crystal surface, and T. Shoji for polishing the crystals superbly. This work was supported in part by a Grant-in Aid for Developmental Scientific Research (No. 12555105) from the Ministry of Education, Science, and Culture of Japan.
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