1.1 Progress in applying THz-wave parametric oscillators
   – Development of a tera-photonics spectroscopic system (TSS) and THz-wave imaging system –

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Abstract
We developed a widely tunable terahertz (THz)-wave parametric oscillator (TPO) that improves on conventional TPOs and is unified with optics. The TPO can be used in various research applications. We also demonstrated a tera-photonics spectroscopic system (TSS) and a THz-wave imaging system with a TPO as applications. We report the first use of a TPO to measure biological samples, such as salmon DNA, bovine albumin, bovine γ-globulin, and cytochrome-c from horse heart using TSS. Moreover, by THz-wave imaging we obtained a clear image of dielectric materials embedded in an opaque envelope. Different images at 1.6 and 1.8 THz and a differential image between two frequencies could be derived.

1. Introduction
Recently, widely tunable THz-wave sources have attracted significant interest in THz-wave applications, such as molecular spectroscopy, materials science, solid state physics, biological research, and gas tracing. Specifically, a THz-wave parametric oscillator (TPO) [1-4] has several features that are superior to those of other THz-wave sources, such as small size, wide tunability, high temporal and spatial coherence, high conversion efficiency, and room-temperature operation. Therefore, such TPOs will be used in a wide range of THz-wave applications.

TPOs are based on stimulated polariton scattering in a nonlinear LiNbO$_3$ crystal pumped by a Q-switched Nd:YAG laser. In the polariton scattering process, the noncollinear phase-matching condition, $k_p = k_T + k_i$, and the conservation of energy, $\omega_p = \omega_T + \omega_i$, are satisfied (p = pump, i = idler, T = THz-wave). Therefore, continuous frequency tuning can be achieved by slightly varying the phase-matching angle between the idler and pump beam. Practically, the angle is varied from 1 to 3 degrees, which corresponds to a frequency range from about 1 to 3 THz. Although precise control of the angle is needed, the tuning method is simple and the reproducibility of the tuning characteristics was confirmed using a precisely rotating stage. Consequently, TPOs can be handled easily and are suitable as practical THz-wave sources for various applications.

This report describes a new THz-wave parametric oscillator that improves on the conventional TPO and which could be used with THz-wave optics, the optics for monitoring the idler beam, and so on. We also demonstrate a THz-wave spectroscopic system and THz-wave imaging system as applications of the TPO. The THz-wave region is spectroscopically significant for scientific and commercial purposes, since it contains a large number of molecular rotational absorption lines. Therefore, spectroscopic and imaging systems in the THz-wave region are important basic analytical tools. We gave careful consideration to ease of handling, so anyone can operate the TSS easily. A THz-wave imaging system was also developed by adding a two-dimensional mechanical scan device to the TSS.

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2. Terahertz-wave parametric oscillator (TPO)

Figure 1 shows the experimental setup and a photograph of the TPO. This TPO was developed for THz-wave research, including collaborations with internal and external organizations. The TPO consisted of a 5mol% MgO-doped LiNbO₃ nonlinear optical crystal, two mirrors, a precise rotation stage, and the optics. The MgO-doped LiNbO₃ crystal was 65 × 4 × 5 mm long × wide × high. The end-surfaces were cut parallel, polished, and coated to make them anti-reflective. The crystal was placed inside the Fabry-Perot cavity, which consisted of two mirrors. Mirror M₁ on the pump beam input side was a half-area coated HR mirror so that the pump beam could be coupled into the cavity. Mirror M₂ was full-area HR coated. The pump beam was reflected by M₂ and coupled out of the cavity. The reflectivity of these mirrors exceeded 98%. Part of the generated idler beam was extracted from mirror M₂, folded by a reflector, and coupled to an optical fiber. Since the mirrors, reflector, and fiber were placed on the same plate mounted on a precise rotation stage (Harmonic Drive Systems LFI-80-6, minimum step angle: 0.06 deg.), the path of the idler was constant and always led to the fiber. Therefore, the idler beam could be monitored outside the system in the middle of tuning operation. The pump laser was a Q-switched Nd:YAG laser that operated at a 50 Hz repetition rate. Its radiation had typical pulse energy of 30 mJ/pulse and 25 ns pulse duration.

The THz-wave was extracted from an arrayed Si prism coupler [5]. Si prisms, which were used due to avoid total internal reflection of the THz-wave in the crystal (n ≈ 5), were fixed to a holder. The nonlinear crystal was attached to the Si prisms. This holder was simpler than the usual holder, so that the crystal could be changed easily when it suffered optical damage.

The THz-wave radiation was divided into two paths at a beam splitter, dividing the THz-wave output in a 3 to 1 ratio to ports 1 and 2, respectively. Figure 2 shows the THz-wave output energy at a pump input energy of 30 mJ/pulse. A tunable range of roughly 1 to 2.1 THz was obtained. A THz-wave output exceeding 0.01 pJ/pulse was obtained in the tuning range. A maximum energy of 7 and 2 pJ/pulse was obtained from the signal and reference ports.
respectively. Using a 4K–Si bolometer, the output energy is sufficient for various measurements with a good signal-to-noise ratio. This TPO is manufactured by PAX Corporation.

3. Tera-photonics Spectroscopy System (TSS)

A tera-photonics spectroscopy system was developed that allowed easy THz-wave spectroscopy using our various THz-wave sources, which include a TPO, injection-seeded THz-wave parametric generator, injection-seeded PPLN optical parametric generator, and so on. Figure 3 shows the setup of the TSS using a TPO. The TSS consisted of a Q-switched Nd:YAG laser, TPO, 4K-Si bolometer, signal-processing circuit, and computer. The construction of the TPO was described in the previous chapter.

The THz-wave extracted from the TPO was divided into reference and signal paths by a standing wire-grid beam splitter. We used an Si bolometer in which two detectors were embedded to measure the reference and signal THz-waves independently and cancel out the output fluctuation. The signals from the Si bolometer were sent to an analog signal-processing circuit, which was operated in synchrony with the repetition frequency of the pump laser, which was 50 Hz in this study. The maximum frequency is up to 1 kHz. The data were transferred to the computer through a peak-hold circuit and a delayed-sampling circuit in the signal-processing circuit. Frequency tuning and data acquisition were controlled by the computer using the Labview intermediate program. We designed the signal-processing circuit and the algorithm of the Labview program, which are manufactured by Ryowa Electronics Corporation.

Recent biological research using applied THz-waves has focused on the low frequency collective vibrational modes that occur in proteins and DNA, since these modes may provide information about the conformational state of biomolecules [6]. We also think that our widely tunable THz-wave source will play an important role in biological and medical research. We have started measuring the THz-wave spectra of typical biological samples using the TSS. Figures 4 (a)-(d) show the spectra of salmon DNA, bovine albumin, bovine γ-globulin, and horse heart cytochrome-c, respectively. The DNA and cytochrome-c were obtained from Wako
Pure Chemical Industries, Ltd. (cord# 047-22491 and cord# 031-13561, respectively). The albumin and γ-globulin were obtained from SIGMA (cord# G7516 and cord# A7638, respectively). These samples were powders and were put in a holder that consisted of two membranes. The characteristics of the holder were cancelled out by measuring an empty holder in advance. In general, the transmittance decreased as the wavenumber increased. Although we did not observe significant absorption lines in these specimens, we will continue our spectroscopic investigations of biological samples.

4. THz-wave imaging using a TPO

Recently, THz-wave imaging has been extensively studied using pulsed THz radiation generated by femtosecond laser pulses and photoconductive dipole antennas [7,8]. The potential of THz imaging, which has a better resolution than microwave imaging, has attracted much attention in applied research. THz-wave imaging using a TPO is expected to extend the range of THz-waves used, since images at different frequencies, and differential images between the transmittances at two frequencies, can be obtained.

Figure 5 shows the experimental setup for our THz-wave imaging. The fundamental configuration of the optics was the same as that of the TSS. In addition, two TPX lenses (f = 30 mm) were placed on the signal path. The THz wave was focused on the imaging target, which was scanned by an x-z stage up to 20 × 20 mm. The frequency tuning, data acquisition, and x-z stage were controlled by a computer via a Labview program, which was an improvement over the TSS control program.

In these measurements, the targets were a T-shaped low-density polyethylene plate and a P-shaped Teflon plate. The polyethylene and Teflon were 170 µm and 1.5 mm thick, respectively. Figure 6 shows the transmittance characteristics of these materials. The low-density polyethylene had a periodic change in transmittance because of an etalon effect.
However, the Teflon had an almost constant transmittance. Figure 7(a)-(c) shows THz-wave images at 1.6 and 1.8 THz and the differential image between two frequencies, respectively. Both the “T” and “P” were emphasized at 1.6 THz, while only the “P” was emphasized at 1.8 THz and only the “T” was emphasized in the differential image. Consequently, different information can be obtained using our THz-wave imaging method with a TPO.

5. Summary

We developed a unified and improved widely tunable TPO. We obtained a tuning range from roughly 1 to 2.1 THz and a maximum output of 7 and 2 pJ/pulse from the signal and reference ports, respectively. Using a 4K-Si bolometer, the TPO can be used for various applications. We also demonstrated TSS and THz-wave imaging using a TPO. In TSS, we report the first use of a TPO to measure biological samples. Moreover, we successfully obtained a clear image of targets embedded in an opaque envelope. Images at 1.6 and 1.8 THz, and a differential image between the two frequencies were obtained.
References