Tunable terahertz-wave generation from DAST crystal by dual signal-wave parametric oscillation of periodically poled lithium niobate

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The first known demonstration of tunable terahertz-wave generation by difference-frequency generation of dual signal-wave quasi-phase-matched optical parametric oscillation was performed with periodically poled LiNbO$_3$ (PPLN) with a series of gratings. An organic ionic salt, 4-dimethylamino-N-methyl-4-stilbazolium-tosylate (DAST), was used as a nonlinear crystal. A compact terahertz-wave source resulted, and changing the temperature of the PPLN permitted the wavelength to be varied from 120 to 160 $\mu$m. The wavelength could be tuned from 100 to 700 $\mu$m by proper selection of combinations of periodically poled gratings. © 2000 Optical Society of America

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The generation of widely tunable coherent terahertz-(THz-)wave radiation is of great interest for a variety of applications in basic and applied physics, communications, and life sciences. The invention of the laser stimulated significant research into nonlinear optics, including research into difference-frequency generation (DFG) of tunable submillimeter waves by use of two laser sources. However, the development of the molecular gas submillimeter laser, despite its inferior tunability, ended such research in the mid-1970’s. Since the late 1980’s, the generation and detection of short THz pulses has attracted a great deal of attention in terms of both theory and application. Most studies have utilized the ultrabroad bandwidth characteristics of mode-locked subpicosecond laser pulses. Developments in the study of widely tunable THz-or submillimeter-wave sources, however, have centered on the $p$-Ge laser, the free-electron laser, photo mixing, and optical parametric oscillation.

We applied a dual signal-wave (DSW) quasi-phase-matched (QPM) optical parametric oscillator (OPO) to THz-wave DFG, using periodically poled LiNbO$_3$ (PPLN) with a series of gratings. This type of PPLN has two periods on the pump path and therefore has two energy-conservation and phase-matching conditions such that the two signal waves are collinearly generated at one pump wave. These properties make the DSW QPM OPO promising for use as a compact DFG THz-wave source. An organic ionic salt, 4-dimethylamino-$N$-methyl-4-stilbazolium-tosylate (DAST), was used as a DFG mixer. We have developed a stable, reproducible method of growing large DAST crystals, using the fixed seed crystal method. DAST, which has been patented by Nakanishi et al., has quite large nonlinear and electro-optic coefficients. In addition, it is suitable for high-speed moduation and detection, as well as for THz-wave radiation, because of its low dielectric constant.

Efficient THz short-pulse radiation from DAST from optical rectification with a subpicosecond laser pulse as the source has been reported. Also, tunable THz-wave generation with an electronically tuned Ti:Al$_2$O$_3$ laser as the optical source has been demonstrated. We have also studied the use of DAST as a milliwave receiver.

The experimental setup is illustrated in Fig. 1. The PPLN was fabricated by the electric field poling...
process. Two grating periods (29.3 and 29.5 μm), each of which had an interaction length of 20 mm, for a total length of 40 mm, were used. We placed the crystal in an oven, both to prevent photorefractive effects and to tune the oscillating wavelengths. The OPO cavity consisted of 50-mm-radius concave mirrors separated by 50 mm. The reflectivities of the input and output mirrors at the signal wavelengths were 99% and 80%, respectively. The two idler waves were absorbed into the mirror substrate. The pump source was a Q-switched Nd:YAG laser (wavelength, 1.064 μm; pulse width, 120 ns; repetition rate, 1–5 kHz; energy, 1 mJ/pulse at 1 kHz). The pump beam was focused to 150 μm (1/e² radius) by a lens (f = 200 mm) at the center of the PPLN secondary grating. The output for each signal was typically 100 μJ/pulse, and the polarizations were parallel to each other.

The two signal beams were focused onto an ~0.5-mm spots on the DAST surface (total crystal size, 12 mm × 11 mm × 1 mm) by an f = 100 mm lens. The THz wave generated from the DAST was focused into a 4-K Si bolometer by two parabolic metal mirrors (f = 120 mm). The transmitted signal beams were eliminated by a small mirror. Figure 2 shows examples of wavelength measurements: Fig. 2(a), of oscillating signal waves at a temperature of 150°; Fig. 2(b), of the generated THz wave. The THz wavelength was measured with a scanning Fabry–Perot etalon consisting of two metal-mesh plates (a 65-μm grid). The displacement between two periods was 139 μm, which corresponds directly to the difference frequency between two incident signal wavelengths (1.529 and 1.546 μm). We tuned the signal wavelength interval from 15 to 20 nm by varying the temperature from 100° to 200°, which resulted in THz-wavelength tuning from 120 to 160 μm. Through proper selection of gating periods, it was possible to tune the signal-wave interval from 2 to 22 nm, corresponding to THz-wavelength tuning from 100 to 700 μm. The purity of the THz wave is determined by the signals’ linewidth. A narrower THz linewidth can be obtained by introduction of an injection-seeding technique into the DSW QPM OPO.

In Fig. 3(a) the THz-wave output variation is plotted as a function of the angle between the a axis of the DAST crystal and the signal polarization. The signal polarizations were rotated by a half-wave plate. The maximum and minimum THz-wave outputs were observed when the signal polarizations were parallel to the a and the b axes, respectively, of the DAST crystal. The polarization direction of the THz wave was measured by a wire grid polarizer and was confirmed to be parallel to the a axis of the DAST crystal, as shown in Fig. 3(b). Note that the THz-wave transmission is maximum when the polarization and the wire are perpendicular to each other. The THz-wave polarization was independent of the signal polarization. These results indicate that the d₁₁₁ property of DAST crystals is utilized in generation of THz waves.

Figure 4 shows the output energy characteristics of the THz-wave DFG as a function of the input energy. The maximum THz-wave output was 52 fJ/pulse at a repetition rate of 1 kHz (52 pW on average), with an incident sum signal energy of 270 μJ/pulse. The repetition rate was increased to 5 kHz, which was the upper limit of the response of Si bolometer. It should be added that we have performed DFG experiments with other crystals, such as LiNbO₃, LiTaO₃, KTP, and GaAs, under the same experimental conditions; however, only DAST generated a detectable THz wave.

In conclusion, a compact and widely tunable THz-wave source was realized by the combination of a DAST crystal and a DSW QPM OPO by use of PPLN with a series of gratings. The THz-wave power was 52 pW for a total DFG input power of 270 mW, with
The conversion efficiency could be increased if the less absorptive frequency range of DAST in the sub-THz region \(^24\) and DFG interaction with a longer coherence length were used.\(^{20}\) We have calculated the dependence of the coherence length on input (near-infrared) and output (THz) wavelengths based on the refractive indices\(^24\) of DAST. Under the experimental conditions reported here, the absorption coefficient for the THz wave (2.16 THz; wavelength, 139 \(\mu\)m) and the coherence length were \(\sim 60\ \text{cm}^{-1}\) and \(\sim 0.9\ \text{mm}\), respectively. For 0.5-THz (wavelength, 600 \(\mu\)m) generation from signal inputs near 1.5 \(\mu\)m, the absorption coefficient and the coherence length are \(\sim 10\ \text{cm}^{-1}\) and \(\sim 3\ \text{mm}\), respectively. We are attempting to design a DSW QPM OPO for sub-THz-wave generation.

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