We developed a widely tunable terahertz (THz)-wave source covering the sub-THz frequency by difference frequency generation using a 4-dimethylamino-N'-methyl-4'-stibazolium tosylate (DAST) crystal. Near-infrared waves generated by dual-wavelength injection-seeded $\beta$-BaB$_2$O$_4$ optical parametric generation (is-BBO-OPG) were used for pumping the DAST crystal, which had separated wavelengths in the spectrum with a difference frequency of sub-THz. Furthermore, the non-collinear phase-matching condition was designed to compensate the walk-off effect of the BBO crystal. Consequently, tunable THz-waves from 0.3 to 4 THz were generated by tuning the wavelength of one of the seeding beams. The generated sub-THz-waves were monochromatic ($\Delta \nu < 33 \text{ GHz}$) with a maximum energy of 80 pJ at 0.65 THz.

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An organic nonlinear optical crystal of DAST has low dispersion of the refractive index from the THz to NIR frequencies, which provides various collinear phase-matching conditions. Additionally, it has a high nonlinear coefficient (1010 pm/V at 1300 nm), which yields a high conversion efficiency of DFG. Consequently, the DFG configuration is suitable for tunable and efficient THz-wave generation. As a pumping source for DAST-DFG, dual-wavelength sources with wavelengths of approximately 1300 nm are necessary to satisfy the optimal phase-matching condition.

To generate a tunable dual-wavelength output at around 1300 nm, we applied a BBO crystal. A BBO crystal is generally used because of its wide tunability and high nonlinear conversion efficiency. We applied a type-I phase-matching condition because the effective nonlinear coefficient is larger than that under the type-II phase-matching condition. With wide tolerance in the oscillation spectrum by the cavity mode and the high gain of the nonlinear crystal. Consequently, the dual-wavelength OPO is inappropriate as a pump source for sub-THz-wave generation.

In this paper, we proposed a 1300 nm injection-seeded $\beta$-BaB$_2$O$_4$-OPG (is-BBO-OPG) without a cavity structure to generate spectrally separated wavelengths with difference frequency down to the sub-THz range. Moreover, to improve the conversion efficiency of is-BBO-OPG, an efficient walk-off compensation in the BBO crystal was designed and demonstrated. Using this is-BBO-OPG as a dual-wavelength pumping source for 4-dimethylamino-N'-methyl-4'-stibazolium tosylate (DAST)-DFG, we demonstrated widely tunable THz-wave generation from 0.3 to 4 THz. A wide phase-matching condition as a function of frequency in the BBO and DAST crystals enabled tunability to be controlled merely by tuning the wavelength in one of the injection-seeding beams. Thus, frequency agility was accomplished with no mechanical motion or temperature control of the crystal. Furthermore, the output of is-BBO-OPG had additional peaks in the spectrum, that are expected to induce cascade DFG for highly efficient THz-wave generation, as reported in a previous paper. The present source has potential to extend the tunability to tens of THz by improving the tuning range of injection-seeding beams.
532-nm-wave pumping, an idler wave of 1300–1400 nm is generated, with the cogeneration of a signal wave at about 900 nm. Figures 1(a) and 1(b) depict two types of phase-matching conditions: non-collinear and collinear phase-matching in the BBO crystal, respectively. The vectors \( k_p, k_s, \) and \( k_i \) respectively signify the wave vectors of the pump, idler, and signal beams. \( S_p \) and \( S_i \) respectively represent the Poynting vectors of the pump and idler beams. \( \alpha, \beta, \) and \( \gamma \) respectively denote the angles of the pump, idler, and signal beams measured from the \( c \)-axis. \( \rho \) is the walk-off angle of the pump beam.

The degree of overlap of the two beams is possible in BBO-OPG owing to the non-collinear phase-matching configuration. Figures 1(c) and 1(d) depict calculated tuning curves of non-collinear phase-matching satisfying Eqs. (1) and (2), where the pump wavelength is 532 nm.

In collinear phase-matching, the wave vectors of all three beams are parallel but the Poynting vectors are not parallel. Because of the birefringence, the Poynting vector of the pump beam with extraordinary polarization (e-polarization) is not parallel to that of the idler beam with ordinary polarization (o-polarization). The degree of overlap of the two beams decreases with propagation. Thereby, the optical parametric gain decreases even if phase-matching is satisfied.

Here, the polarization of the signal beam is ordinary. To achieve better overlap between the pump beam and idler beam, the non-collinear phase-matching configuration is applicable. Non-collinear phase-matching is effective for increasing the degree of overlap between these beams by compensating the walk-off effect. As shown in Fig. 1, the design of non-collinear phase-matching in our is-OPG is depicted for parallel Poynting vectors of the pump and idler beams. In this configuration, the walk-off angle (\( \rho \)) and the tilt angle of the idler from the pump beam (\( \beta - \alpha \)) are equal, which allows parallel overlap. For this configuration, the following two equations must be satisfied.

\[
\left( \frac{n_o(\lambda_s)}{\lambda_s} \right)^2 = \left( \frac{n_e(\lambda_p, \alpha)}{\lambda_p} \right)^2 + \left( \frac{n_o(\lambda_i)}{\lambda_i} \right)^2 - 2 \left( \frac{n_e(\lambda_p, \alpha)}{\lambda_p} \right) \left( \frac{n_o(\lambda_i)}{\lambda_i} \right) \cos(\beta - \alpha)
\]

Equation (1) represents non-collinear phase-matching in Type-I BBO. Equation (2) represents the parallel propagation of pump and idler beams owing to the walk-off effect. Using experimental values, the solutions for \( \alpha, \beta, \) and \( \gamma \) are listed in Table I for the pump and idler beam wavelengths of 532 and 1330 nm, respectively. The values for collinear phase-matching are also listed in Table I for comparison. In collinear phase-matching, the deviation angle of the Poynting vector of the pump beam from that of the idler beam (\( \alpha + \rho - \beta \)) is 3.2°, but it is zero in non-collinear phase-matching. The calculated results showed that parallel propagation of the pump and idler beams is possible in BBO-OPG owing to the non-collinear phase-matching configuration. Figures 1(c) and 1(d) depict calculated tuning curves of non-collinear phase-matching satisfying Eqs. (1) and (2), where the pump and idler beams propagate in parallel. The results indicate that a wide tuning range (\( \delta \lambda = 50 \) nm) is possible with a fixed pump angle because the difference in the phase-matching angle is within 0.16° in the tuning range; this value is less than the divergence angle of the pump beam.

The corresponding tuning range of the maximum difference frequency is approximately 8 THz. The tuning curve of collinear phase-matching is also depicted in Figs. 1(c) and 1(d) for comparison.

Table I. Values for non-collinear (NCL) phase-matching configuration calculated from Eqs. (1) and (2) with pump and idler wavelengths of 532 and 1330 nm, respectively. Values for collinear (CL) phase-matching are also listed.

| \( \lambda_{\text{pump}} \) (nm) | \( \alpha \) (deg) | \( \beta \) (deg) | \( \gamma \) (deg) | \( \rho \) (deg) | \( \alpha + \rho - \beta \) (deg) |
|----------------|----------------|----------------|----------------|----------------|----------------|
| NCL 532        | 20.7           | 26.9           | 21.5           | 3.2            | 0.0            |
| CL 1330        | 22.6           | 22.6           | 22.6           | 3.2            | 3.2            |

\[
\tan \beta = \left( \frac{n_e(\lambda_p)}{n_o(\lambda_i, 90^\circ)} \right)^2 \tan \alpha
\]
600 nm focal length. The power intensity at the crystal was 4.8 GW/cm². The incident angle of the pump beam to the crystal was individually adjusted to satisfy the non-collinear or collinear phase-matching condition. To inject seeding beams for the idler beam, two O-band external cavity laser diodes (ECLDs) (6324, New focus; tuning range of λ = 1270–1330 nm; frequency linewidth of <300 kHz) were coupled with the pump beam, so that the injection beams satisfied the non-collinear phase-matching condition and the collinear phase-matching condition, which were measured to compare the performances. The output power of each seeding beam was increased to 6 mW using optical semiconductor amplifiers. The generated idler outputs with a dual wavelength regime were 4.0 and 1.8% under non-collinear and collinear conditions, respectively.

Figures 3(a) and 3(b) show spectra of the OPG outputs without (blue) and with seeding (red) of λ1 = 1323 nm and λ2 = 1326 nm, under the (a) non-collinear phase-matching condition and (b) collinear phase-matching condition. The spectra were measured using a spectral analyzer (ANDO AQ6315A, minimum wavelength resolution of 0.05 nm). In non-collinear phase-matching, the suppression of optical parametric fluorescence by seeding was 8 dB. Efficient seeding produced a higher signal-to-noise ratio of about 30 dB between the peak intensity and residual fluorescence intensity. In collinear phase-matching, the suppression of fluorescence was only about 2 dB and the signal-to-noise ratio was 15 dB. In comparison, the suppression of fluorescence intensity was about twice as high under the non-collinear phase-matching condition than that under the collinear phase-matching condition. The walk-off compensation increased the overlap of pump and seed beams, resulting in the greater suppression of fluorescence intensity. Additionally, the higher signal-to-noise ratio in non-collinear phase-matching is important for efficient THz-wave generation in the latter DFG process. The energy of is-BBO-OPG was measured with a power meter (OPHIR 3A). The CW power of seeding beams was subtracted from the measured energy of is-BBO-OPG. The values of the maximum energy of is-BBO-OPG were, respectively, 0.44 and 0.21 mJ under non-collinear and collinear phase-matching conditions. The conversion efficiencies from the pump to the idler output were 4.0 and 1.8% under non-collinear and collinear conditions, respectively.

Figure 3(c) shows the spectrum of the idler output in non-collinear phase-matching measured with the higher wavelength resolution. The spectrum had five clearly separated peaks equally spaced with a frequency of 0.47 THz, which exactly equals the frequency difference between the two seeding beams. A multiple-wavelength output was measured in the entire tuning range, where the peaks are equally spaced at intervals equivalent to the difference frequency of the two seeding beams. The generation of multiple wavelengths resulted from the four-wave mixing of the two wavelengths, λ1 and λ2, with a high gain of the BBO crystal. The multiple wavelength output is expected to induce the cascade DFG process for highly efficient THz-wave generation.

For the generation of THz-waves, the NIR output of is-BBO-OPG was focused onto a 9 × 9 × 0.8 mm³ DAST crystal by a planoconvex lens with a 100 mm focal length. The polarization of the NIR beam was aligned to the a-axis of the DAST crystal under the type-0 phase-matching condition to make use of the high nonlinearity and broadband tunability. After the NIR beam pumped the DAST crystal, the residual beam was absorbed by the IR filter, which only transmitted the THz-wave. The THz-wave from the DAST crystal was collimated by a Tsurupica lens with a 50 mm focal length. The THz-wave was detected by a 4 K Si bolometer with a black polyethylene film on the window to remove the rest of the IR wave in order to transmit only the THz-wave. A far-infrared filter with a cutoff frequency of 3.6 THz was also inserted to remove the stray light. The output voltage of the Si bolometer was measured using an oscilloscope. During measurements, optical paths of the THz-wave were purged with dry N₂ gas to prevent absorption loss by water vapor in air.

Figure 4(a) shows the bolometer output as a function of the THz-wave frequency. Measurements were done three times
with different dynamic ranges in the oscilloscope, because the THz-wave output changed greatly by two orders of magnitude depending on the frequency. Data of three ranges of frequency were merged for seamless and continuous connection of the graph. Tuning of the THz-wave frequency was achieved by changing the wavelength of one seeding beam in the range of \( \lambda_1 = 1302 \text{ to } 1325 \text{ nm} \) with a resolution of 17 GHz, while the wavelength of the other seeding beam \( \lambda_2 \) was fixed at 1326 nm. The results of the bolometer output showed that the THz-wave output was widely tunable from 0.3 to 4 THz. The maximum energy of sub-THz-waves was 80 pJ at 0.65 THz, corresponding to a peak power of 0.8 W, where the pulse width of 100 ps was inferred from the results of an earlier experiment. The sensitivity of the bolometer was measured using a calibrated pyrometer. The conversion efficiency of energy from NIR to sub-THz-waves was approximately 5 \times 10^{-7}. The highest THz-wave frequency was limited by the cutoff frequency of the mid-infrared filter inserted in the bolometer.

Figure 4(b) shows the wavelength measurement of the THz-wave generated by the scanning Fabry–Perot etalon. The etalon was placed in front of the bolometer. The etalon was formed of two Si plates, where one of the Si plates was set on a mechanically movable stage. The free spectral range (FSR) and finesse of the etalon were estimated to be 75 GHz and 2.3, respectively. The wavelengths of the seeding beams were fixed at 1323 and 1326 nm, respectively. The periodical intensity peaks were measured by increasing the distance between the two Si plates. The distance between two adjoining peaks is 320 \mu m, which is half-the wavelength of the input THz-wave and corresponds to the frequency of 0.47 THz, which matches the difference frequency of the two seeding beams. The frequency linewidth of the sub-THz-wave was estimated to be 33 GHz by fitting analysis of these data. The measured results showed the generated sub-THz-wave to be monochromatic. Therefore in the DFG process, only the nearest pair of peaks in the multiple-wavelength pump beam was efficient for the generation.

In conclusion, we demonstrated widely tunable and monochromatic THz generation by DAST-DFG pumped by the NIR-wave output of is-BBO-OPG. We developed dual-wavelength is-BBO-OPG seeded by two seeding beams with a narrow spectrum. For is-BBO-OPG, we designed non-collinear phase-matching to optimize the overlap of the pump beam and the idler beam. In the designed configuration, efficient seeding and high conversion efficiency were achieved. The is-OPG generated a spectrum with peaks separated by a sub-THz difference frequency. By pumping DAST-DFG by is-OPG, a widely tunable THz-wave of 0.3–4 THz was generated and was fully controlled by the seeding beam wavelength. The output THz-wave was monochromatic. The non-collinear phase-matching configuration proposed herein is expected to be applicable for other crystals possessing a walk-off effect, such as KTP, KTA, KDP, BiBo, LBO, CLBO, LiTaO₃, and LiNbO₃. Moreover, the technique of generating clearly separated wavelengths in the developed NIR source is expected to be applicable to other pumping sources for exciting nonlinear crystals reported in the literature related to THz-wave generation, such as organic crystals of BNA, DASC, BDAS-TP, DSTM, and OHI and semiconductor crystals of GaP, GaSe, ZnSe, and ZnTe. The developed DAST-DFG system pumped by is-BBO-OPG is anticipated to be a promising system for a widely tunable THz-wave source covering the sub-THz frequency region, and should be applicable for nondestructive imaging, remote sensing, or the calibration of sensitivity in novel sub-THz-wave detectors.

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