Terahertz Wave Generation from GaP with Continuous Wave and Pulse Pumping in the 1–1.2 μm Region

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The terahertz (THz) region, which lies between the microwave and infrared regions, offers a wealth of untapped potential. We generated widely frequency-tunable coherent THz waves from GaP crystal pumped using continuous-wave (CW) semiconductor lasers and compared it with pulse pumping using Q-switched high-power lasers at 1–1.2 μm . THz wave generation was based on difference-frequency generation via the excitation of phonon-polaritons in GaP. CW THz waves were generated from GaP by enhancing the power density of the pumping light from semiconductor lasers. The power and phase-matched condition for THz wave generation are discussed with respect to the pumping method compared to pulse pumping. Semiconductor lasers have light power stability with a narrow linewidth. Therefore, CW THz waves can be used as a light source in high-resolution THz spectroscopy, as well as in multichannel communication. [doi:10.2320/matertrans.48.980]

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1. Introduction

Many terahertz (THz) wave sources have been developed recently,^{1–13)} and THz technologies have increased in many fields, including the solid-state physics of acoustic phonons, optical phonons, impurity energy levels, defect energy levels, and sub-band transitions in superlattices.^{14–16)} The THz properties of materials are important for both investigating fundamental physical properties and for developing THz applications such as generators, detectors, and amplifiers in the THz region.

In 1963, Nishizawa^{1,2)} proposed THz wave generation from compound crystals via the excitation of phonons or molecular vibrations. In 1983, the semiconductor Raman laser was realized, which relied on the longitudinal optical phonon (LO phonon) mode of a GaP crystal.⁵⁾ The output power of the Raman laser was increased by a phonon enhancement effect within waveguide-structured GaP.^{17,18)} Using a GaP Raman laser, a 12.1-THz wave was generated at a power of 3 W.^{6,7)} Recently, wide-frequency-tunable high-power THz waves have been generated from GaP by pumping with a Q-switched YAG laser and an optical parametric oscillator (OPO) at 1 μm .^{19–21)} THz waves were generated using difference frequency generation (DFG) via the excitation of phonon-polaritons based on the Raman effect in GaP. THz waves were also obtained using Cr:forsterite lasers at 1.2–1.3 μm ,²²⁾ which have the merits of simple structure, easy maintenance, and low cost compared to pumping using YAG and an OPO. Here, the Cr:forsterite lasers were pumped with a double-pulse Q-switched YAG laser.

Most THz waves are generated by pumping with Q-switched laser pulses. Pulsed THz waves have limited

applications, whereas continuous-wave (CW) THz waves are more desirable for the expansion of THz technologies to applications such as broadband telecommunications and imaging technologies. Imaging using CW THz waves will enable *in situ* security screening techniques. Until now, the CW THz waves generation has been developed by using a photoconductive antenna.^{23,24)} CW THz waves pumped using semiconductor lasers have such a narrow linewidth, *i.e.*, on a MHz order, that we can obtain high-resolution THz spectra rapidly. Semiconductor lasers are stable at light intensities, and spectrometers using CW THz waves do not need prolonged measurements with much repetition. Because THz wave sources using a Q-switched pulsed laser have repetition rates as low as 10 Hz with large power fluctuations, spectrometers using Q-switched pulsed lasers can produce low-noise spectra only after averaging many measurements.

Here, we describe the generation of widely tunable CW THz waves from GaP based on laser diode (LD) pumping and compare it with pulse pumping using Q-switched high-power pulses at 1–1.2 μm . The power and phase-matched condition of THz wave generation is discussed with respect to the pumping method. The two beams used for CW THz wave generation were an external cavity laser diode (ECLD) and an LD-pumped Nd:YAG laser combined with an ytterbium-doped fiber amplifier (FA).

2. Experimental

The experimental setup for CW THz waves generation is shown in Fig. 1. The pump and signal lasers used for DFG in GaP were an ECLD and a laser diode-pumped YAG laser (1.064 μm) with ytterbium-doped FA, respectively. The wavelength of the ECLD can be varied with a linewidth of 2 MHz. The power from the ECLD and FA was 0.24 and 3.6 W, respectively, before the beams were incident on the GaP crystal. The diameter of the amplified 1.064- μm beam was about 300 μm using a lens with $f = 1200$ mm. The ECLD beam was elliptical in cross-section (5×2.5 mm)

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Fig. 1 Schematic of the optical setup used for continuous-wave THz wave generation from GaP using semiconductor ECLD and LD-pumped YAG lasers.

before it was focused with a lens with $f = 600$ mm. The waist of the focused beam was also elliptical and measured $150 \times 300 \mu\text{m}$.

A GaP crystal was cut into a rectangle measuring 10 mm in the $\langle 110 \rangle$ direction and 3 mm in the $\langle 001 \rangle$ direction. The pump beam from the ECLD was combined with the signal beam from the FA using a polarizing cube beam splitter placed on a rotating platform on top of a linear stage, which automatically produced a very small angle between the two beams to fulfill the phase-matching (PM) condition and achieve spatial overlap of the incoming beams. The incident beams were almost parallel to the $\langle 110 \rangle$ crystal direction of the GaP based on Raman effect.²⁵⁾

The THz wave output was collected using a polyethylene lens and detected with a liquid-helium-cooled Si bolometer. The Si bolometer was placed in the path of the generated THz waves, which leave the GaP crystal output face at 0.70 – 0.79 rad to the surface normal. Black polyethylene film and 0.8 mm thick crystalline quartz and garnet powder were used to filter out near-IR radiation, which limited the detectable frequency below 3 THz. The bolometer signal was measured using a lock-in amplifier.

While, the pulsed THz waves have been generated from the GaP using YAG and OPO, or Cr:forsterite lasers as $1 \mu\text{m}$ and $1.2 \mu\text{m}$ pulse pumping, respectively. The pulse generator pumped at $1 \mu\text{m}$ region uses a Q-switched Nd:YAG laser with 10 Hz repetition rate and a $\beta\text{-BaB}_2\text{O}_4$ (BBO)-based OPO pumped by using the YAG laser.^{19–21)} Its pulse width and line width were 11 ns and 90 MHz for the $1.064\text{-}\mu\text{m}$ fundamental beam of the YAG laser, and 6 ns and less than 6 GHz for the 1.035 – $1.064 \mu\text{m}$ tunable beam of the OPO laser, respectively. The energies of the YAG and OPO were 3 mJ with 3 mm diameter. The other pulse generator pumped at $1.2 \mu\text{m}$ region uses two Cr:Forsterite lasers.²²⁾ The Cr:F lasers are pumped by a Q-switched YAG laser with a 10 Hz repetition rate. The wavelength of the pump beam was fixed to 1210 nm. The linewidth was less than 0.01 nm with an etalon and 0.1 nm without an etalon. The wavelength of the signal beam was varied between 1210 and 1250 nm by tuning the prism dispersion and linewidth was below 0.1 nm. The pulse durations and input powers of the both beams were 20 – 30 ns and 4 mJ (1 mm diameter), respectively. The THz wave

Fig. 2 Frequency dependence of the maximum THz wave output power from various semiconductor crystals. Note that the pump (OPO) and signal (YAG) energies were 3 mJ before the beams were incident on the crystal: continuous-wave GaP (●), pulse GaP (△), GaSe (□), ZnGeP₂ (◇).

energy was collected using parabolic reflectors and detected using a liquid helium-cooled Si bolometer or a pyroelectric deuterium triglycine sulfate (DTGS) detector. A Ge plate was used to filter out near-infrared beams and attenuate the THz wave. The other components of the THz spectrometers and their operation were almost the same as the CW pumping.

3. Results and Discussion

Continuous-wave (CW) THz wave generation should be possible from GaP by enhancing the power density of the pumping light. When the pumping power is limited, the THz wave output power (P_{THz}) can be increased in inverse proportion to the beam spot size, S , based on the following equation:

$$P_{\text{THz}} = \frac{A}{S} \cdot P_1 \cdot P_2 \quad (1)$$

where A is the coefficient for generating THz waves from GaP with pulse pumping at $1 \mu\text{m}$ under noncollinear PM conditions. Figure 2 shows the frequency dependence of the maximum THz wave output power from different semiconductors, where the pulse energies of the YAG and OPO were both attenuated to 3 mJ before they were incident on the crystals. The pulsed THz wave output power from GaP remained high and stable at approximately 0.6 nJ/pulse over a wide THz frequency range. The THz wave output power increased linearly with the pump and signal beam energy. It increased to 9 nJ/pulse in a GaP THz wave generator when the respective pump and signal energies of the Cr:forsterite lasers were 11.4 and 11.6 mJ with a spot size of 1 mm. A is estimated as $0.4 \times 10^{-13} \text{ W}^{-1} \text{ cm}^2$ from the results of pulse pumping at $1 \mu\text{m}$.^{19–21)} In this CW THz wave generator from GaP, the spatial overlap of the cross-sectional areas of the pump and signal beams, S , was $3.5 \times 10^{-4} \text{ cm}^2$. The effective powers of the pump and signal beams, P_1 and P_2 , were 0.24 and 1.8 W, respectively. Therefore, the expected THz power was 50 pW around 2 THz if the optical alignment were perfect. THz waves were generated over the range from 0.69 to 2.74 THz. The frequency dependence of the CW THz wave output power has the same tendency for the relative power spectrum as that with pulse pumping.

Fig. 3 Dependence of the PM angle on the THz wave frequency for the maximum continuous-wave THz wave output power for each frequency. The symbols $\square$ and $\circ$ represent the result of pulse pumping at 1 and 1.2 μm , respectively. The solid and dashed lines represent the calculated relationship for pumping at the two wavelengths.

Figure 3 illustrates the relationship between the THz wave frequency and θ_{in}^{ext} at which the maximum output power was obtained, where θ_{in}^{ext} is the external angle between the pump and signal beams outside the GaP crystal. Noncollinear PM for THz wave generation was satisfied (Fig. 3), including the results at 1 and 1.2 μm .^{19–22} The PM condition for CW THz wave generation was similar to the result for pulse pumping.^{19–21} Because the two beams are noncollinear, the internal angle inside the GaP crystal, θ_{in} , is given by

$$\theta_{in} \approx \frac{\theta_{in}^{ext}}{n_S} \quad (2)$$

where n_S is the refractive index of GaP at 1.064 μm (3.105). The θ_{in} was calculated as 0.017 rad at 2 THz. Therefore, the two near-IR beams overlapped sufficiently to interact with each other in the GaP. The polarization of the pump and signal beams was adjusted to be in the $\langle 001 \rangle$ and $\langle 1\bar{1}0 \rangle$ directions, respectively. The angle between the pump and signal light beams (θ_{in}) was calculated using eq. (3) and matched the experimental results well. It corresponded to the dispersion curve of the phonon-polariton branch of GaP.²⁶

$$\frac{\theta_{in}}{v} = \sqrt{\frac{2(\Delta q/q)(n_I^2/n_L \cdot n_S)}{v_L \cdot v_S}} \quad (3)$$

where, v_L , v_S , and v are the frequencies of the pump, signal, and THz waves, respectively; $(\Delta q/q)$ is the relative deviation of phonon-polaritons in GaP; and n_I , n_L , and n_S are the refractive indices of the THz wave, pump, and signal lights, respectively. The θ_{in} increases with the wavelength of the pump and signal beams, as seen in pulse pumping. Therefore, the output direction of the THz wave depends on the wavelength of the pump and signal beams, as well as the THz frequency.

The PM angle bandwidth for half the maximum power at a fixed frequency was plotted as a function of the THz wave frequency (Fig. 4). The curves show the calculated angle bandwidth for each frequency bandwidth (400–600 GHz) for the fixed θ_{in}^{ext} using eq. (3). The calculated angle increased with the frequency bandwidth (Fig. 4). The experimental

Fig. 4 Frequency dependence of the PM angle bandwidth for half the maximum power at a fixed THz wave frequency. The curves show the calculated angle bandwidth for each frequency bandwidth (400–600 GHz) for a fixed angle between the pump and signal beams using eq. (3).

Fig. 5 THz wave output power plotted as a function of the THz wave frequency when $\theta_{in}^{ext} = 3.67 \times 10^{-3}$ rad. The dashed line is a visual guide for the experimental points.

value was the same as that for a frequency bandwidth of 600 GHz. The THz wave output power was plotted as a function of the THz wave frequency for $\theta_{in}^{ext} = 3.67 \times 10^{-3}$ rad (Fig. 5). The frequency bandwidth for half the maximum power was 600 GHz. Experimentally, there was good agreement between the angle and frequency bandwidth calculated using eq. (3). This indicates that eq. (3), which is based on the dispersion curve of the polariton branch of GaP, is well explained for THz wave generation by the excitation of phonon-polaritons with CW, as well as with pulse pumping.

4. Conclusion

Widely frequency-tunable single-frequency continuous-wave (CW) THz waves were generated based on the Raman effect via the excitation of phonon-polaritons in GaP by semiconductor lasers and compared with pulse pumping sources consisting of a Q-switched Nd:YAG laser, OPO, and Cr:forsterite lasers. The frequency dependence of the THz wave output power was similar for the relative power spectrum comparing CW and pulse pumping. The PM condition of THz wave generation was determined by the

wavelength of the pump and signal lights regardless of the pumping method, where the PM angle between the light beams increased with the wavelength. The PM angle bandwidth was confirmed experimentally and interpreted theoretically based on the dispersion of the polariton branch of GaP in which THz waves are generated by excitation of the phonon-polariton. The frequency bandwidth for half the maximum power was 600 GHz at a fixed PM angle.

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