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GaP THz wave generator and THz spectrometer using Cr:Forsterite lasers

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We have developed a type of THz wave generator that uses Cr:Forsterite lasers as the pump and signal sources for difference frequency generation in GaP (Cr:F source system). We confirmed the generation of THz waves in the frequency range from 0.3 to 7.5 THz, which is just similar to that obtained using the THz wave generator previously developed utilizing yttrium aluminum garnet and optical parametric oscillator (OPO) lasers (OPO source system). A peak output power of 100 mW was obtained from 1.2 to 5 THz when the power of the two input beams was 3 mJ each, similar to the OPO source system. A wide measurable frequency range from below 0.6 THz to over 6 THz was obtained by using the Cr:F source system as the light source of a spectrometer, which has the merits of simple structure, easy maintenance, and low cost compared with the OPO source system. Although the linewidth of the Cr:F source system is greater than that of the OPO source system, the THz spectrometer still has sufficient resolution for measuring solids or liquids at room temperature. © *2005 American Institute of Physics.* [DOI: 10.1063/1.2140223]

In 1963, Nishizawa[1,2] proposed the generation of terahertz (THz) waves via the resonance of phonons and molecular vibrations to fill the frequency gap between the microwave and optical bands, just after the realization of a semiconductor laser, which Nishizawa also proposed and patented.[3] Loudon presented a similar proposal for THz wave generation using a uniaxial crystal[4] and Yarborough *et al.* observed THz wave generation via dielectrics ($LiNbO_3$).[5] Following the development of a semiconductor GaP Raman laser,[6] Nishizawa and Suto generated a 12-THz wave with a peak power as high as 3 W using a GaP Raman oscillator containing a GaAs mixing crystal.[7] Kawase *et al.* improved the efficiency of THz wave generation based on the Nishizawa-Yarborough method by introducing a grating coupler on the $LiNbO_3$.[8] Moreover, they demonstrated frequency-sweepable, high-power THz wave generation using the parametric amplification effect of lattice vibrations in the frequency range up to 2.5 THz using $LiNbO_3$.[9–11] We have developed higher-power THz wave generation from GaP tunable over wider frequencies.[12–14] The principle of our method is difference frequency generation (DFG) via excitation of the phonon-polariton mode under small-angle noncollinear phase-matching conditions. Using an yttrium aluminum garnet (YAG) laser and an optical parametric oscillator laser as the pump and signal sources, respectively, a tunable frequency range of 0.3–7.4 THz available by changing the crystal length from 2.6 to 20 mm, with a linewidth of 3.2 GHz, and a maximum peak power of 100 mW at 2.5 THz was obtained when the pump and signal energies were 3 mJ. When the crystal length was increased to 20 mm, the maximum power was increased to 300 mW, but the frequency range was reduced to below 5 THz. We constructed an automatic THz spectrometer using GaP frequency-tunable THz wave generators and reported the spectra of various biomolecules, including saccharides, nucleosides, and nucleotides, demonstrating the merits of our system for spectrometric analysis.[15–18] In this article, we report the characteristics of frequency-tunable THz wave generation from GaP using Cr:Forsterite (Cr:F) lasers as the pump and signal source for DFG. We present a THz spectrometer system and an example of measurements.

Figure 1 shows a schematic of the experimental setup used for DFG of THz waves in a GaP crystal using Cr:F lasers as the source (Cr:F source). Both the pump and signal beams were delivered from two channel Cr:F lasers (LOTIS TII) pumped with a double-pulse Q-switched Nd:YAG laser at 1064 nm. A Cr:F laser generates a 1.2–1.3 μm wavelength beam utilizing the Cr levels in a Forsterite crystal. Cr:F source systems can be simple and small because they use direct excitation by the fundamental wave of a YAG laser. The repetition rate was 10 Hz and the pulse timing of each channel could be controlled independently on a nanosecond order. This is important because the delay time changes with the wavelength. The wavelength of the pump

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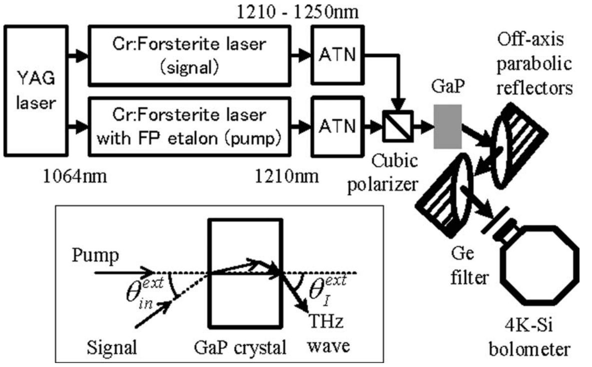

FIG. 1. Experimental setup for DFG of THz waves in GaP crystal using Cr: lasers.

beam was fixed to 1210 nm and the linewidth was below 0.01 nm with an étalon or below 0.1 nm without an étalon. The wavelength of the signal beam was variable from 1210 to 1250 nm and the linewidth was below 0.1 nm. Each beam was 1 mm in diameter with a 22 ns pulse length, and more than 6 mJ pulse energy per pulse. The polarization of the pump and signal beams was adjusted vertically and horizontally, respectively. The two beams were combined at a very small angle to fulfill the phase-matching condition using a cubic polarizer. The undoped semi-insulating GaP crystal was 5 or 10 mm long in the ⟨110⟩ direction and 3 mm thick in the ⟨001⟩ direction. Both the input and output face of the crystal were coated with Al_2O_3 after mechanical polishing and chemical etching. The THz waves generated were collected with a pair of off-axis parabolic reflectors and detected using a 4 K Si bolometer. A Ge plate was used as a filter to cut off near-IR light.

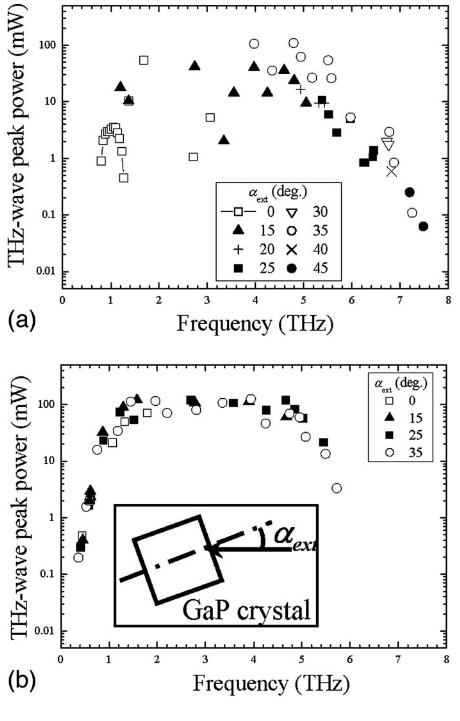

FIG. 2. Frequency dependence of the maximum THz wave output power at various θ_{in}^{ext} for (a) 5-mm-long and (b) 10-mm-long GaP crystals.

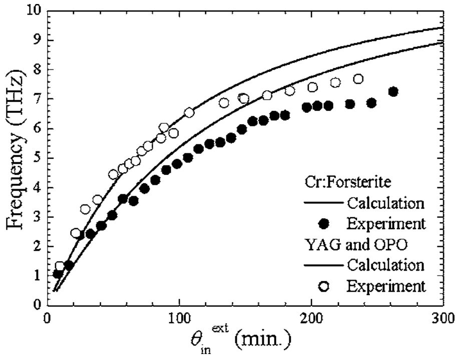

FIG. 3. Relationship between θ_{in}^{ext} and the THz wave frequency giving the maximum THz power. An output angle of α was selected for the optimum condition.

Figure 2 shows the frequency dependence of the maximum THz wave output peak power at various θ_{in}^{ext}, where θ_{in}^{ext} is the external angle between the pump and signal beams outside the GaP crystal. The GaP crystal lengths were (a) 5 or (b) 10 mm long. The two beams were attenuated to 3 mJ before incidence. As mentioned below, the incident angle α_{ext} of the pump and signal beams from the normal of the GaP input surface had to be increased to avoid total reflection of the THz wave at the output surface. The α_{ext} was selected at each frequency to obtain the maximum output power. THz generation was observed at frequencies from 0.6 to 7.5 THz for a 5-mm-long crystal. The maximum THz wave output power was 150 mW for a 5-mm-long crystal. These values are of the same order as those obtained using another type of THz generator that uses YAG and OPO lasers as the source (OPO source system). One of the notable feature of Cr:F source system is its high output power, which increases to 1.5 W for a 10-mm-long crystal without causing surface damage, when the pump and signal energies are 11.4 and 11.6 mJ, respectively. Figure 3 plots the relationship between θ_{in}^{ext} and the generated THz wave frequency at which the maximum power was obtained. The incident angle α_{ext} was also changed at each point to obtain the maximum power. The solid curve shows θ_{in}^{ext} as a function of THz wave frequency calculated using the dispersion relationship of phonon-polaritons in GaP. Although the calculation concurs with the measurements at lower frequencies, the discrepancy increases at higher frequencies. This may be due mainly to the increase in α_{ext} to avoid total reflection. In Fig. 3, the same relationship is plotted for the GaP THz generator utilizing an OPO laser as the pump source. The THz wave generator with the Cr: lasers has a larger θ_{in}^{ext} than that with the OPO laser because the lower frequencies of the incident beams, which cause a larger refractive index of the THz wave, need a larger angle to fulfill the phase-matching condition to generate a THz wave at the same frequency.

Figure 4 shows a schematic diagram of an automatic measurement system for transmission spectroscopy using a Cr:F source system as a THz light source. The equipment used for THz transmittance spectroscopy is similar to the OPO source system reported in detail in our previous papers.[15,16] The phase-matching condition was fulfilled automatically with rotating and linear stages to control the beam angle and an actuator to control the wavelength of the Cr:F laser. The GaP crystal was placed on a rotating stage and the incident angle α_{ext} was adjusted to prevent total reflection. The maximum signal-to-noise (S/N) ratios (output power/

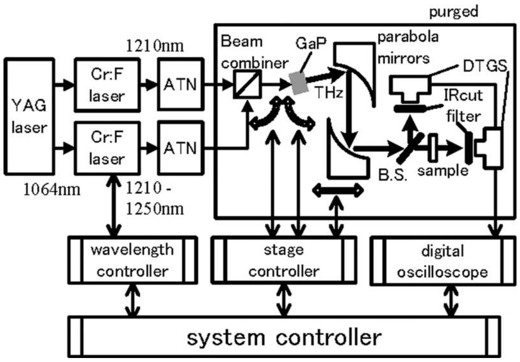

FIG. 4. Schematic diagram of a THz spectrometer using Cr:F lasers.

detector noise) were about 5000 for a deuterium triglycine sulfate (DTGS) detector and over 1 000 000 for a 4 K bolometer. The spectrometer systems were built using the double-beam method to eliminate the effect of THz power fluctuation. Actually, THz power fluctuations are 25% and 10% for OPO and Cr:F source systems, respectively, so that the S/N ratio is better for the Cr:F source system by a factor of 2.5 even when the double-beam method is used. Pyroelectric DTGS detectors operated at room temperature were used as THz wave detectors. Black polyethylene filters were used to cut off near-IR light. The entire THz wave path was purged with dry nitrogen or dry air to eliminate water vapor absorption. The spectrometer measured about $120 \times 100 \times 100$ cm, including the source lasers and power supplies, while the OPO source system has a large size of $300 \times 100 \times 100$ cm. The linewidth of the THz wave is 30 GHz, while that for the OPO source system is 1.5 to ~3.2 GHz. Note that 30-GHz resolution is sufficient for room temperature observations of THz spectra in solids or liquids.

Figure 5 shows transmittance spectra of α-D-glucose measured with the Cr:F source system (solid line) in 15-GHz steps with a linewidth better than 30 GHz (1 cm^{-1}), and with the OPO source system (dashed line) in 10-GHz steps with a linewidth of 1.5 GHz (0.1 cm^{-1}), measured at 294 K by sweeping the 0.5 to ~5.6 THz (23.3 to ~187 cm^{-1}) frequency region. The spectra were obtained by averaging 32 pulses of the DTGS outputs at each frequency. Crystalline glucose purchased from Tokyo Kasei Kogyo was milled with polyethylene (PE) powder and pressed into 1-nm-thick pellets (20 mm diam) under a pressure of 2000 kg. The glucose concentration was 5 wt % in an approximately 300-mg PE pellet. The sample pellets were shaped into a wedge in order to prevent THz wave resonance via an étalon effect. The main absorption bands observed with the OPO source system were also observed with the Cr:F source system. Note that the absorption band at 1.49 THz is sharp compared to the other bands. The full width at half maximum (FWHM) of the band was obtained as 62 GHz using the Cr:F source system and 50 GHz using the OPO source system.

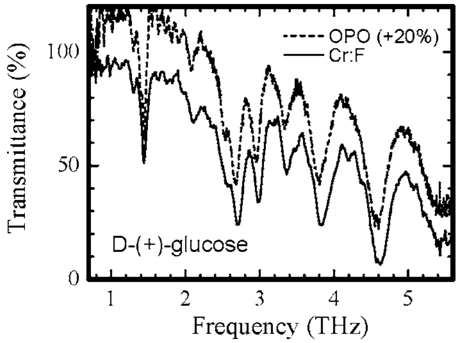

FIG. 5. Transmittance spectra of α-D-glucose measured using the Cr:F source THz spectrometer system.

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