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THz transmittance measurements of nucleobases and related molecules in the 0.4- to 5.8-THz region using a GaP THz wave generator

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Abstract

An automatic measurement system for terahertz (THz) transmission spectroscopy based on difference-frequency generation (DFG) of widely tunable coherent THz waves via excitation of a phonon–polariton mode in GaP was constructed and improved to give it a wider frequency measurement range and four times greater S/N ratio by using the double-beam method. In this paper, we present spectroscopic measurements of nucleobases, nucleosides, deoxynucleosides, and nucleotides, all of which are the components of RNA and DNA molecules, in the range of 0.4–5.8 THz in order to compare their characteristic features in the wider terahertz frequency region.

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1. Introduction

In 1963, Nishizawa [1,2] predicted coherent terahertz electromagnetic wave generation via the resonance of phonons and molecular vibrations. Following this proposal, Nishizawa and Suto [3–5] realized a semiconductor Raman laser using a GaP crystal, and a terahertz (THz) wave with a frequency of 12 THz was generated from a GaP Raman laser containing a GaAs mixing crystal [6]. Loudon [7,8] also proposed terahertz wave generation, but based on a uniaxial crystal. Nishizawa promoted the development of THz wave generation via the resonance of phonons and

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proposed that a wavelength-tunable THz wave would be applicable to detect and treat of cancers [9]. At the same time, under Nishizawa's guidance, Kawase and Ito [10,11] realized high-power frequency-tunable terahertz wave generation from $LiNbO_3$. Its narrow line width (100 MHz) enabled the spectral measurement of sharp water vapor lines with high resolution. In addition, Tanabe et al. [12–15] reported high-power frequency-tunable terahertz wave generation via the excitation of phonon–polaritons in GaP, with a maximum peak power as high as 4.8 nJ/pulse (800 mW) at 2.5 THz, and frequency-tunable in the range of 0.5 to 7 THz without missing frequencies.

Recently, we constructed a GaP terahertz wave generator with automatic scanning control in the range of 0.8–5.4 THz and made spectral measurements of terahertz vibrations of sugars, including glucose, deoxyglucose, fructose, and sucrose [16]. THz spectra of some sugar molecules have been measured [17] using terahertz time domain spectroscopy (THz-TDS) based on photoconductive switches [18,19], which were developed by several groups [20–40]. Terahertz spectra measured using a GaP frequency-tunable THz generator have revealed high frequency portions (above 3.5 THz) of resonance peaks that are hidden using conventional THz-TDS. The time domain THz wave radiation mentioned above has a pulse form. By contrast, the frequency-tunable THz wave obtained using the phonon–polariton mode of GaP or $LiNbO_3$ has tunable quasi-CW frequency characteristics.

Based on the THz-TDS, THz waves were applied for imaging of biological tissues and electronics [41–47]. Jepsen et al. [48–52] measured THz spectra of biomolecules using THz-TDS. Recently, Kawase et al. [53] applied THz imaging to the non-destructive detection of illicit drugs using spectral fingerprints and a $LiNbO_3$ tunable THz wave source.

In this paper, we present spectroscopic measurements of important biomolecules in the range of 0.4–5.8 THz using an improved GaP tunable-frequency THz wave source. These biomolecules are the essential components of RNA and DNA: the five nucleobases (adenine, guanine, cytosine, uracil, and thymine) and their nucleosides, deoxynucleosides, and nucleotides. These spectra differ markedly within this THz region, facilitating the identification and discrimination of biomolecules. Only limited data have been available on the resonance behavior of these molecules in the THz frequency region. Thus, the first step in the spectroscopic analysis requires the construction of a database of THz spectra of such kinds of biomolecules.

Our spectrometer was improved by introducing a sample and reference beam path configuration (double-beam method) to decrease the effect of THz power fluctuations, as shown in Fig. 1. Single-frequency THz wave radiation from the GaP THz wave generator has a 10-ns pulse form with a repetition frequency of 10 Hz. Consequently, the main source of noise in the spectrometer arises from the fluctuation in the THz intensity. This

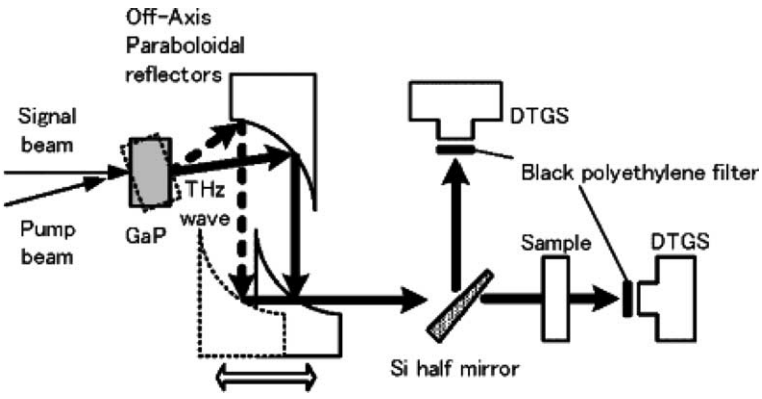

Fig. 1. Configuration of the THz transmission spectroscopy measurement system.