Comparative study of by near-field heterodyne transient grating and continuously variable spatial frequency transient grating methods for measurements of terahertz reflection responses

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Abstract. We measured terahertz reflection responses utilizing a propagating phonon polariton wave, which was generated and detected by two different methods: the near-field heterodyne transient grating and the continuously variable spatial frequency transient grating method. The obtained results were compared for the purpose of clarifying which of both methods is better for measurements of terahertz reflection responses. The phonon polariton wave is propagated and reflected at a ferroelectric crystal edge. From the viewpoint of the separation between the excited and reflected phonon polariton waves, the latter method is better for measurements of reflection responses.

1. Introduction
In recent years, research utilizing electromagnetic waves in a frequency range of 0.3-10 terahertz (THz: 10^12 Hz) has progressed considerably.¹ Since terahertz frequencies are located in between those of radio and light waves, terahertz waves feature high permeability, similar to radio waves and easy handling using optics, similar to light waves. Many applications have been proposed and various technologies have been developed using the terahertz electromagnetic wave.²⁻⁴ For example, various molecules, such as those of vitamins, sugars, medicines, and agricultural chemicals, with similar structures can be distinguished using the characteristic oscillation bands of each molecule.⁵⁻⁸ Furthermore, molecular interactions of sugar and amino acids were investigated, and crystal polymorphisms were classified based on the fact that terahertz waves are sensitive to small changes in van der Waals interactions.⁵⁻⁸

In terahertz spectroscopy, femtosecond optical pulses are used for generating terahertz fields that are projected from sources into free space and detected after transmission or reflection by a sample using a separate receiver.⁹,¹² However, an integrated platform that supports terahertz generation,
propagation, transmission and characterization would be more versatile and convenient. For this purpose, a method using optically generated phonon polariton (PP) waves offer strong prospects.

A PP wave is a coupled mode of an electromagnetic wave and a polar lattice vibration, and in nonlinear crystals such as LiNbO$_3$ (LN) and LiTaO$_3$, it can be excited through the impulsive stimulated Raman scattering (ISRS) mechanism. In practice, a wavelength selective PP wave can be generated by using a transient grating optical configuration and PP waves at an arbitrary frequency can be obtained because the dispersion relation has been well established experimentally and theoretically. Since PP can propagate at light like speed on the spatial range of millimeters to centimeters, it can be used as a terahertz light source within crystal using reflected responses.

Since a conventional transient grating (TG) setup is very large and complicated for the integrating terahertz platform, we have adopted the near-field heterodyne TG (NF-HD-TG) setup for generation and detection of a PP wave, which features a very simple and compact optical setup. This is important for the integration of terahertz PP waves into a small crystal because the PP waves can travel for centimeters at most. Furthermore, we have developed a continuously variable spatial frequency TG (CVSF-TG) setup, which is superior in its tunability of frequency compared to the NF-HD-TG method, and this method was actually used for the reflection measurements of the PP wave.

In this study, we measured the reflection responses of PP waves by both methods and compared the results for the purpose of clarifying which of both is better for measurements of the reflection responses.

2. Experimental
The optical configurations of NF-HD-TG method and CVSF-TG method are shown in Figure 1. The optical setup is almost the same between the NF-HD-TG method (a) and CVSF-TG method (b). The difference is that the frequency is tuned by a glass plate with many transmission gratings with different grating

![Figure 1. A schematic representation of optical setup of (a) NF-HD-TG method and (b) CVSF-TG method for the generation and detection of a PP wave.](image)

![Figure 2. Typical responses of a PP wave measured by (a) NF-HD-TG method and (b) CVSF-TG method with a frequency of 0.44 THz and 0.7 THz, respectively.](image)
spacings in the NF-HD-TG method, while the motion of a single transmission grating can give a different grating spacing in the CVSF-TG method.

In the NF-HD-TG technique, a coherent pump light is incident onto a transmission grating, and the light intensity profile close to the transmission grating on the side opposite the incident light has striped patterns. When a ferroelectric crystal is placed at the optical fringe pattern of light, PP waves are generated by way of the ISRS process. When a PP wave is excited, the refractive index is changed according to the “intensity of the polarization”. When a probe light is incident in the same manner as the pump light, it is diffracted both by the transmission grating (reference) and by the polariton grating (signal) and the signal response of the diffracted beam mixed with the reference beam is detected by heterodyne detection.

On the other hand, in the CVSF-TG method, when coherent light is sent through a transmission grating after being passed through a lens, and a ferroelectric crystal is placed at a distance d behind the focal position, the light intensity profile of the crystal has an image pattern of the transmission grating itself; the image size is reducible or expandable to D/d of the original grating. (D: distance between the grating and focal plane) This image can be used as an optical fringe pattern, and its spacing can be continuously controlled by moving the transmission grating in the light propagation direction, namely by varying D. The probe light transmits through the same lens and transmission grating and overlaps on the crystal where the PP wave is initially excited. When a part of the grating image intensity transmitted through a pinhole is measured, the field intensity of the original grating image overlapped with the modulated field caused by the PP wave. As a result, the two fields are mixed and detected using heterodyne detection. The PP wave excited by each method propagates almost parallel to the x axis and the wave is reflected at the crystal edge. After reflection, it comes back along the x axis, and the reflected response is measured by each method again.

3. Result and Discussions
Figures 2. (a) and (b) show typical responses of a PP wave measured by the NF-HD-TG method and the CVSF-TG method for signals with a frequency of 0.44 THz and 0.7 THz, respectively. The vibration response in the time interval between 0 and 15 ps is due to the excited PP wave. Usually, the peak response around 0 ps is observed due to the optical Kerr effect.

Figures 3. (a) and (b) show reflected responses of a PP wave measured by the NF-HD-TG and the CVSF-TG method, respectively. The dependence on the distance between the excited position and the edge of the crystal (L) is shown for a frequency of (a) 0.44 THz and (b) 0.6 THz, respectively. Figure 3. (a) is vertically expanded to clearly show the reflected PP waves, and the excited PP waves are not within the window. We could successfully observe small vibrational responses emerging on different times depending on L for both the methods. As L increased, we observed

Figure 3. The PP wave responses for different distances of the excitation position to crystal edge (L) by (a) NF-HD-TG method and (b) CVSF-TG method, respectively.
these responses at a later time. From these results, the observed small vibration was confirmed to be due to the reflected PP wave at the crystal edge. In order to measure the reflection of a PP wave, it is necessary to detect the reflected PP wave response with a sufficiently high S/N ratio and to separate the reflected PP wave response from the excited PP wave response. In case of the NF-HD-TG method, L should be shorter than 2.5 mm for detection with a high S/N ratio because the PP wave gradually decays due to damping as it propagates. However, L should be longer than 1.5 mm; otherwise, the excited PP waveform overlaps with the reflected PP waveform and a pure reflected waveform cannot be obtained. Therefore, L should be shorter than 2.5 mm and longer than 1.5 mm. In case of the CVSFG-TG method, L should be shorter than 2.5 mm for high S/N ratio detection because the PP wave gradually decays due to damping as it propagates and L should be longer than 0.25 mm. Therefore, L should be shorter than 2.5 mm and longer than 0.25 mm.

In the NF-HD-TG method, L should be longer to separate the reflected PP wave response from the excited PP wave response compared with the CVSFG-TG method. Since the PP wave is attenuated by propagation for a longer distance, the CVSFG-TG method is superior to the NF-HD-TG method from the view point of S/N ratio. Moreover, the CVSFG-TG method is superior in the frequency tunability because only a single grating is used in this method, while many gratings with different spacings are necessary for the NF-HD-TG method; it cannot be applied if the necessary terahertz frequency corresponds with a wavelength that lies in between two available grating spacings.

4. Conclusion
We have measured terahertz reflection responses utilizing a propagating PP wave and we have compared the results for the purpose of clarifying which of two implemented methods, the NF-HD-TG method or the CVSFG-TG method, is better for measurements of reflection responses. In each method, we obtained the reflected PP responses, but from viewpoint of propagating distance of PP and S/N ratio, the CVSFG-TG method is superior to the NF-HD-TG method. Moreover, the CVSFG-TG method is superior in the frequency tunability to the NF-HD-TG method.

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