Boson Peak Evaluation of Sodosilicate Glass by Terahertz Time-Domain Spectroscopy

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***Abstract*—Terahertz time-domain spectroscopy was performed to investigate the different compositions of sodosilicate glasses around the boson peak (BP) frequency range at temperatures from 13 K to 295 K. We successfully detected the BP behavior at low temperatures. Additionally, we observed the dependence of the temperature and composition of these materials on the BP behavior. Particularly, the BP was clearly observed at 1.09 THz at the lowest temperature for NS4 glass. Furthermore, the BP exhibited broadening as the temperature increased.**

I. INTRODUCTION

DISORDERED solids are characterized by a lack of long-range structural order, which influences their vibrational dynamics. The appearance of an excess of vibrational modes over the Debye level at the terahertz (THz) frequencies is called the boson peak (BP). Additionally, the BP appears in the spectrum of $g(\nu)/\nu^2$, where $g(\nu)$ is the vibrational density of states (VDOS). To determine the origin of the BP, several experimental and theoretical studies have been performed over the past several decades. For a disordered system, $\alpha(\nu)$ and $g(\nu)$ obey the relationship $\alpha(\nu) = C_{\mathrm{IR}}(\nu) \cdot g(\nu)$, where $\alpha(\nu)$ is the absorption coefficient, and $C_{\mathrm{IR}}(\nu)$ is the infrared (IR) light-vibration coupling coefficient. Consequently, we demonstrated that terahertz time-domain spectroscopy (THz-TDS) is suitable for detecting the BP in the spectrum of $\alpha(\nu)/\nu^2$ [1-4].

Sodosilicate is a typical alkali silicate. For understanding the behavior of alkali oxides in silicate, previous researchers calculated the VDOS over a wide range using molecular dynamics [5,6]. However, several issues remain unresolved in the low-frequency region (i.e., the origin of the BP). In this study, different compositions of sodosilicate glasses, with the chemical formula $x\mathrm{Na_2O}\bullet(1\text{-}x)\mathrm{SiO_2}$ (x = 0.2, 0.25, and 0.33, hereafter denoted as NS4, NS3, and NS2, respectively) were selected for the THz-TDS characterization to detect and compare the BP behavior at low temperatures.

As a time-domain technique, THz-TDS can simultaneously yield both the amplitude and phase of a THz electric field [7]. THz-TDS uses a setup with low-temperature-grown GaAs photoconductive antennas for both the emission and detection (RT-10000, Tochigi Nikon Co.), covering the frequency range of 0.25–4.5 THz. The spectroscopic characterization was performed using the standard transmission configuration for the temperature-dependent measurements. The temperature was varied from 13 K to 295 K using a liquid-helium flow cryostat system (Helitran LT-3B, Advanced Research Systems) [8-12].

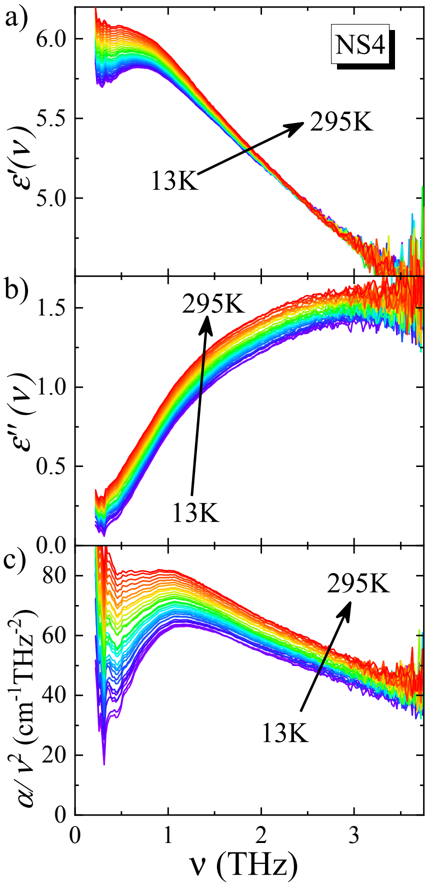

Fig. 1. Temperature dependence of (a) the real parts, $\varepsilon'(\nu)$, and (b) imaginary parts, $\varepsilon''(\nu)$, of the complex dielectric constants and (c) the plot $\alpha(\nu)/\nu^2$ vs. ν of NS4 glass during the heating process from 13 K to 295 K measured via THz-TDS.

II. RESULTS

Fig. 1 (a) and (b) show the temperature dependence of the real ($\varepsilon'(\nu)$) and imaginary ($\varepsilon''(\nu)$) parts of the complex dielectric constant spectrum of NS4 glass from 13 K to 295 K. The values of $\varepsilon'(\nu)$ and $\varepsilon''(\nu)$ increase in the temperature range from 13 K to 295 K. In Fig. 1 (a), a peak is clearly observed around 0.75 THz at the low temperature. This peak becomes broad and blurry as the temperature increases. This behavior results from the appearance of several relaxation processes in the low-

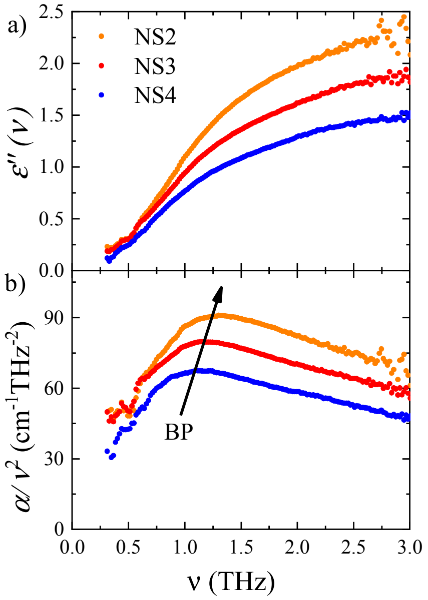

Fig. 2. (a) Imaginary part, $\varepsilon''(\nu)$, of the complex dielectric constants and (b) the plot of $\alpha(\nu)/\nu^2$ vs ν for NS2, NS3 and NS4 glasses at 13 K measured by the THz-TDS.

frequency region and is related to the liquid-glass transition [13]. Furthermore, Fig. 1 (b) shows an inflection point, which is a characteristic of the BP behavior [2]. To provide a direct observation of the BP behavior, Fig. 1 (c) shows the plot of $\alpha(\nu)/\nu^2$ vs. the frequency (ν) of NS4. The BP is clearly observed at 1.09 THz at 13 K. As the temperature increases, the BP shape becomes broad. Finally, it becomes indiscernible at 295 K.

Fig. 2 (a) shows the variation in $\varepsilon''(\nu)$ for NS2, NS3, and NS4 glasses at 13 K. As the Na_2O concentration increases, the value of $\varepsilon''(\nu)$ increases. This trend also leads to an increase in $\alpha(\nu)$. For comparison of the BP behavior, the plots of $\alpha(\nu)/\nu^2$ vs. ν for NS2, NS3, and NS4 glasses is shown in Fig. 2 (b). As the Na_2O concentration increases, the BP frequency shifts to a higher value, and the intensity increases.

III. SUMMARY

We successfully detected the BP at 1.09, 1.13, and 1.30 THz for NS4, NS3, and NS2 glasses, respectively, at low temperatures, using THz-TDS. Specifically, in the case of NS4, the BP shape becomes indistinct as the temperature increases. However, with the increase in Na_2O concentration, the BP frequency shifts to a higher value, and the intensity of the BP increases.

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