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TERAHERTZ AND INFRARED SPECTROSCOPIC STUDY ON DIELECTRIC PROPERTIES OF $Bi_2(Zn_{1/3}Nb_{2/3})_2O_7$ FOR MICROWAVE APPLICATION

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Dielectric properties of $Bi_2(Zn_{1/3}NB_{2/3})_2O_7$ (BiZN) ceramic materials have been studied using terahertz (THz) and Fourier transform infrared (FTIR) spectroscopies. Real part of dielectric constant (ε_1) characterized by FTIR spectroscopy is around $(\varepsilon_1)_{IR}\cong 60$ in low frequency regime (f<100 cm^{-1}) and decreases dramatically in the vicinity of lattice vibrational resonance frequencies or wavenumbers, approaching $(\varepsilon_1)_{IR}\cong$ 2□4 in high frequency regimes (f>1000 cm^{-1}). Real part of dielectric constant (ε_1) characterized by THz spectroscopy is a constant value in 0.1□0.8 THz regime, $(\varepsilon_1)_{THz}\cong 68$, which is essentially the same as the ε_1 value obtained by conventional Hakki-Coleman microwave method. These results reveal that there is no lattice vibrational resonance occurring between THz and microwave (or millimeter wave) frequency regimes. Our results indicate that the low frequency dielectric response of BiZN microwave materials is mainly contributed by the ionic polarization.

Keywords: microwave ceramics; dielectric properties; FTIR and THz

INTRODUCTION

Microwave dielectrics, Bi_2O_3-ZnO-Nb_2O_5 series materials,[1-3] exhibit marvelous microwave properties, and applications of these materials as microwave devices have been extensively investigated. Fourier transform infrared (FTIR)[4] and terahertz (THz)[5,6] spectroscopies are powerful tools for characterizing the response of materials to higher frequency electromagnetic waves. Most of the lattice vibrational phenomena for ceramics, which influence the microwave dielectric behavior of the materials, occur in these frequency regimes. Moreover, no test-fixture is needed, which minimizes the measuring errors. Fourier Transform Infrared (FTIR) and Terahertz (THz) spectroscopies are thus adopted in this study for investigating the microwave dielectric properties of $Bi_2(Zn_{1/3}Nb_{2/3})_2O_7$ (BiZN) materials.

EXPERIMENTAL PROCEDURE

BiZN bulk ceramic was synthesized by using conventional mixed-oxide process, and the microwave dielectric properties of the materials were measured by a Hakki-Coleman[7] method using H. P. 8722 network analyzer. In terahertz spectroscopy, a mode-locked Ti:Sapphire laser of 770 nm wavelength with 600 mW average output power generates ~100 fs pulses at a 76 MHz repetition rate. The experimental setup can measure the loss and dispersion properties of materials up to 1.0 THz range. In infrared spectroscopy, far-infrared reflectance was measured using a Fourier transform infrared spectrometer PERKIN-ELMER System 2000, which possesses a resolution of 4 cm^{-1}. The reflectance measurements were performed over a wide frequency region between 200–10000 cm^{-1} using the deposited aluminum as a reference.

RESULTS AND DISCUSSIONS

The BiZN bulk ceramic material is polycrystalline pyrochlore [Fig. 1(a)]. The typical far infrared reflection spectrum for the BiZN materials is illustrated in Fig. 1(b) (solid curve). To derive the dielectric properties of the materials from the FTIR spectra, Kramer-Krönig model was used[8]. The dielectric properties of the BiZN ceramics thus obtained are shown in Figs. 2(a) and 2(b) (dotted curves) for real part (ε_1) and imaginary part (ε_2) of the dielectric constant, respectively. The ε_2–λ^{-1} curve reveals clearly the occurrence of two lattice vibrational resonances (f_1=320 cm^{-1} and f_2=480 cm^{-1}). The ε_1–λ^{-1} curve indicates that the real part of dielectric constant ($\varepsilon_1 \cong 30$) is large in low frequency regime (f<100 cm^{-1}), and is small ($\varepsilon_1 \cong 2$–4) in high frequency regime (f>1000 cm^{-1}). The abrupt decrease in ε_1 value is attributed to the occurrence of lattice vibrational resonances.

Note that Fig. 1(b) strongly infers the existence of additional resonance in the vicinity of f_0=200 cm^{-1}. To take the f_0-resonance peak into account, the reflectance curve $R(\omega)$ is fitted using Lorentz model[8]. As illustrated in Fig. 1(b), the reflectance $R(\omega)$ modeled by using 3 lattice vibrational resonances (f_0, f_1 and f_2, dash-dotted curve) fits the measured $R(\omega)$–λ^{-1} curve much better than that modeled by using only 2 lattice vibrational resonances (f_1 and f_2, dotted curves). The low frequency (f<100 cm^{-1}) dielectric constant modeled by 3 resonances [dash-dotted curves, Figs. 2(a) and 2(b)] is raised to $(\varepsilon_1)_{IR} \cong 60$, and the dielectric constant in $f \gg f_1$ regime remains at the same small values as compared to those fitted by 2 lattice vibrational resonances [dotted curves, Figs.

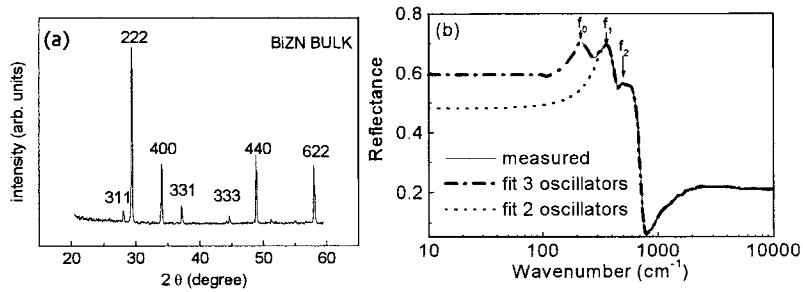

Fig. 1 (a) X-ray diffraction pattern and (b) infrared reflectance spectra of $Bi_2(Zn_{1/3}Nb_{2/3})_2O_7$ bulk material.

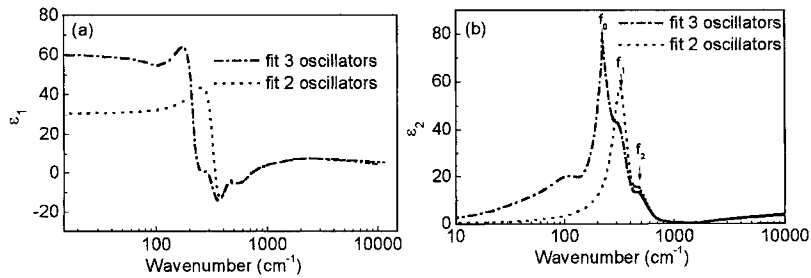

Fig. 2 (a) Real part and (b) imaginary part of dielectric constant fitted by 2 and 3 Lorentz oscillators in FTIR spectra for BiZN bulk materials.

2(a) and 2(b)].

To facilitate the comparison, the dielectric response of the BiZN materials in lower frequency regime (f=0.1□0.8 THz) was measured by terahertz (THz) spectroscope. The complex transmission function $t(\omega)$ due to the presence of BiZN material, was obtained after the original THz waveform was Fourier transformed [Fig. 3(a)]. The complex dielectric constant, $\varepsilon^*=\varepsilon_1+i\varepsilon_2$, was then solved numerically.

Fig. 3(b) reveals that the real part of dielectric constant (ε_1) of BiZN materials varies insignificantly, whereas the imaginary part or loss of dielectric constant (ε_2) increases monotonously with frequency in 0.1□ 0.8 THz regime. The real part of dielectric constant (ε_1) of BiZN materials, around 68, is essentially the same as that measured at 1 GHz by Hakki-Coleman method, inferring that no lattice vibrational resonance phenomenon occurs in between microwave (~1 GHz) and terahertz (1 THz) frequency regimes.

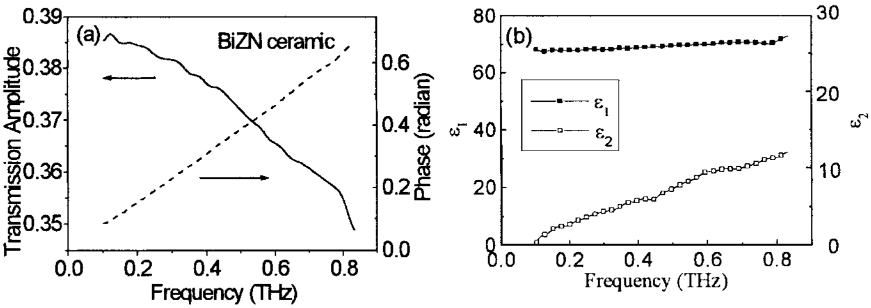

Fig. 3 (a) The transmission amplitude and phase derived from time domain terahertz (THz) waveforms through the BiZN ceramic sample. (b) Real part and imaginary part (loss) of the dielectric constant obtained by using THz spectroscopy.

It should be noted that the real part of dielectric constant (ε_l) evaluated from FTIR spectra using 3-resonance model is markedly closer to that measured in terahertz regime than using 2-resonance model. These results reveal that full spectrum information is needed in order to understand the dielectric response of the materials. THz ▯ FTIR spectroscopies clearly indicate that, for BiZN materials, the lattice vibrational resonances which influence the dielectric properties of the materials, occur in infrared regime. Our results indicate that the low frequency dielectric response of the materials is mainly contributed by the ionic polarization, which is related to vibration of lattices and the high frequency dielectric behavior is predominated by the electronic polarization, which is independent of lattice vibration.

CONCLUSION

Dielectric properties of $Bi_2(Zn_{1/3}NB_{2/3})_2O_7$ ceramic materials were characterized from terahertz to infrared frequency regime using terahertz (THz) and Fourier transform infrared (FTIR) spectroscopies. The real part of dielectric constant (ε_1) analyzed by FTIR spectroscopy is around $(\varepsilon_1)_{IR}\cong 60$ in low frequency regime (f<100 cm^{-1}) while the real part of dielectric constant (ε_1) characterized by THz spectroscopy is about $(\varepsilon_1)_{THz}\cong 68$ in 0.1–0.8 THz regime, which is essentially the same as the ε_1 value obtained by conventional Hakki-Coleman method. These results reveal that there is no lattice vibrational resonance occurring between THz and microwave (or millimeter wave) frequency regime. THz–FTIR spectroscopies provide complete information about the dielectric response of BiZN materials.

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