Journal of the European Ceramic Society 21 (2001) 2787–2790

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Terahertz spectroscopic technique for characterizing the microwave dielectric properties of $Ba(Mg_{1/3}Ta_{2/3})O_3$ materials

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Abstract

We have measured the complex index of refraction of $Ba(Mg_{1/3}Ta_{2/3})O_3$, BMT, ceramics using coherent terahertz (THz) time domain spectroscopy. The dielectric properties of BMT ceramics, which were prepared via a two-step mixed oxide process or a hot isostatic press (HIP) process in 0.1–0.6 THz regime were studied. The real part of the index of refraction of BMT prepared via a two-step process was about 5.04, whereas that of BMT materials prepared via a HIP process was around 4.97. These values are similar to the real part of the index of BMT ceramics in the microwave range ($n = 5.0$). Hipped BMT materials exhibit much larger power loss and hence lower Q-factor than two-step mixed oxide processed BMT in 0.1–0.6 THz regime.

Keywords: $Ba(Mg_{1/3}Ta_{2/3})O_3$; Dielectric losses; Microwave ceramics; THz spectroscopy

1. Introduction

Coherent terahertz (THz) time domain spectroscopy, with its high sensitivity and wide bandwidth, has become a powerful tool for studying the electromagnetic properties of materials in the region of the electromagnetic spectrum that is not easily accessible.[1-4] This novel technique has been applied to a number of material systems for the measurement of the conductivity of doped semiconductors,[5] the absorption coefficient and refractive index of dielectrics,[1] the superconducting band gap in niobium films,[6] the surface impedance of $YBa_2Cu_3O_{7-\delta}$ superconducting films,[7] and the conductivity of $YBa_2Cu_3O_{7-\delta}$ superconducting films.[2,3] With this technique, both phase and amplitude information can be obtained directly from the measurements, so that the real and imaginary parts of the index of refraction, or conductivity, of the samples under study can be determined without using Kramers–Kronig analysis.

Perovskite $Ba(Mg_{1/3}Ta_{2/3})O_3$ (BMT) ceramics possess the highest Q-factor ($\sim$35000 at 10 GHz) among microwave dielectric materials, with low temperature coefficient of resonance frequency ($\tau_f = 5$ ppm per °C) and a large microwave dielectric constant ($\varepsilon_r = 25$). Therefore, these materials have great potential for device applications.[8–11] However, the existence of impurity phase or other defects can increase the dielectric loss. It was found that the losses depend on the method of preparation.[12] While the influence of processing parameters and detail microstructure on the dielectric properties of BMT materials has been thoroughly characterized in microwave regime, the correlation of these characteristics in higher frequency regime is still not well understood. The discrepancy is due to lack of suitable characterization method.

BMT ceramics have been investigated using backward-wave-oscillator (BWO) and FTIR transmission spectroscopy in the 0.2–3.0 THz range.[12,13] In this letter, we used terahertz (THz) transmission spectroscopy for evaluation of the dielectric properties of BMT ceramics, which were prepared via a two-step process or a hot isostatic press (HIP) process in a broadband frequency range from 0.1 to 0.6 THz.

2. Experimental setup and preparation

Fig. 1 schematically shows the experimental setup for THz transmission measurements. A cw Ar^+ laser pumped, mode-locked Ti:sapphire laser generates 100 fs optical pulses at 790 nm with a 76 MHz repetition rate. A large-aperture photoconducting antenna is used as a THz transmitter in our THz spectrometer. The transmitter

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0955-2219/01/$ - see front matter
PII: S0955-2219(01)00364-8

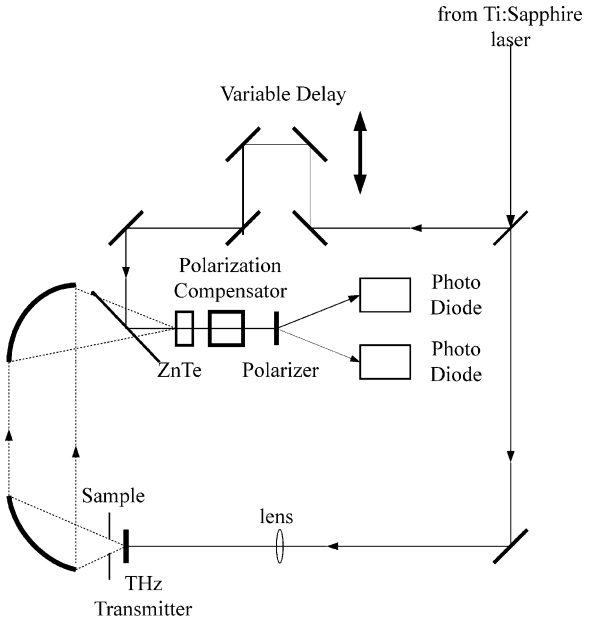

Fig. 1. A schematic setup for THz spectrometer.

consists of a semi-insulating GaAs wafer with two electrodes separated by about 500 μm, on which AuGe is deposited. The bias field is 2 kV per cm and the average optical power on the transmitter is 400 mW. Illuminating the area between the two electrodes with focused ultrashort laser pulses produces synchronous bursts of THz transient. The major fraction of the laser-generated burst of THz transient is emitted into the GaAs substrate in a cone normal to the interface. The scattered THz transient is collected and reflected by an off-axis paraboloidal mirror. When a THz transient is focused onto the electro-optic crystal, the index of refraction is modulated via the Pockels effect. A second optical pulse is delayed with respect to the pump signal by a variable amount, and probes the change in the index of refraction by passing through the crystal. The ellipticity modulation of the probe pulse can be polarization analyzed to obtain information on both the amplitude and phase of the THz field. In this experiment, a 1-mm-thick (110) ZnTe nonlinear crystal is used as a Pockels cell to detect the THz pulses.

The BMT ceramics were prepared via two different processes, a two-step mixed oxide process and a hot isostatic pressing (HIP) process. The two-step process can be described as: (i) the $MgTa_2O_5$ powders were first prepared by a mixed oxide process, (ii) the $MgTa_2O_5$ powders were then mixed with $BaCO_3$ in a 1:3 molar ratio for nominal composition BMT and then calcined at 1200 °C. Pure perovskite BMT powders were then pelletized and were sintered at 1550 °C for 4 h. Thus, obtained materials are designated as "two-step BMT". More detailed discussion on BMT preparation is available in Ref. 14. The HIP process can be described as follow: the $Ba(Mg_{1/3}Ta_{2/3})O_3$, BMT, powders were first prepared by a mixed oxide process. The BMT samples were first conventionally sintered at 1580 °C in air and the hot isostatic pressed in 99.9% Ar atmosphere using a Mo chamber. The temperature of Hipping chamber was raised to 1300 °C with a rate of 7 °C/min, under a pressure of 1500 kg per cm^2. The temperature was held at 1300 °C for 1 h and then was lowered to room temperature with a rate of 16 °C/min, which the argon pressure gradually reduced to 1000 kg per cm^2. The pressure was released at room temperature. Thus obtained materials are designated as Hipped BMT.

3. Results and discussion

Both the two-step processed BMT and Hipped BMT are perovskites with high degree of ordering, as shown in Fig. 2. The dielectric constant and quality factor of the BMT materials measured at 10 GHz are $K=25$ and $Q\times f=60{,}000$ GHz. In THz experiment, reference data is first obtained without the sample between the transmitter and the receiver. Subsequently, a sample was introduced in the path of the THz beam and a second set of data is taken. Fig. 3 shows the typical time domain THz waveform. The spectra of THz transient can be obtained by applying a fast Fourier transform (FFT) to the time domain waveform. Dividing the spectra obtained with a sample by the spectra obtained without a sample yields the transmission function $t(\omega)$ of the sample.

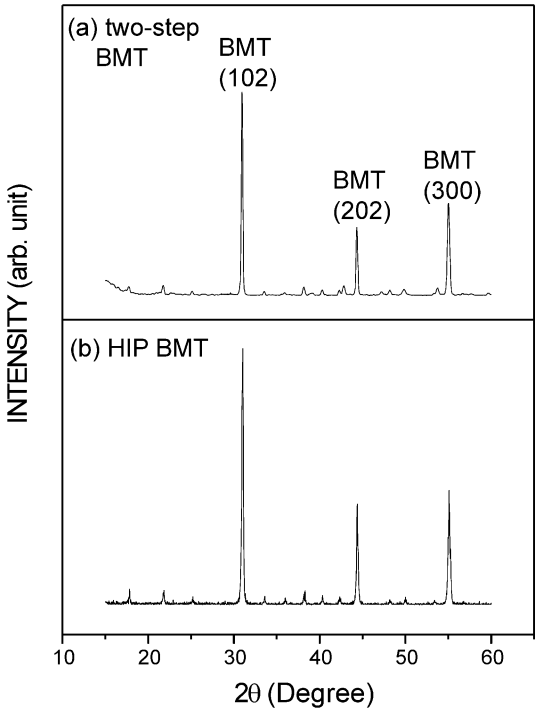

Fig. 2. The X-ray patterns of BMT: (a) two-step processed BMT and (b) hot isostatic pressed BMT.

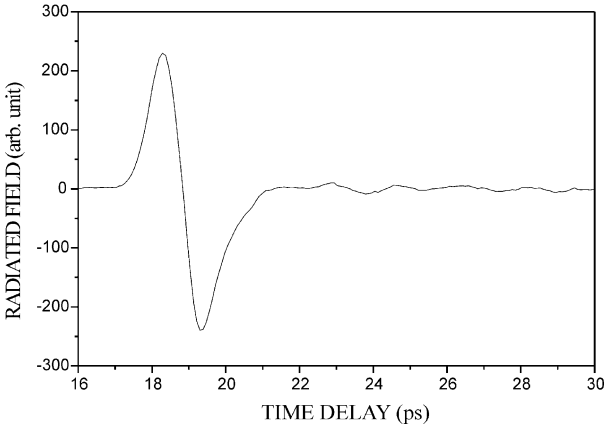

Fig. 3. The typical time domain THz transients.

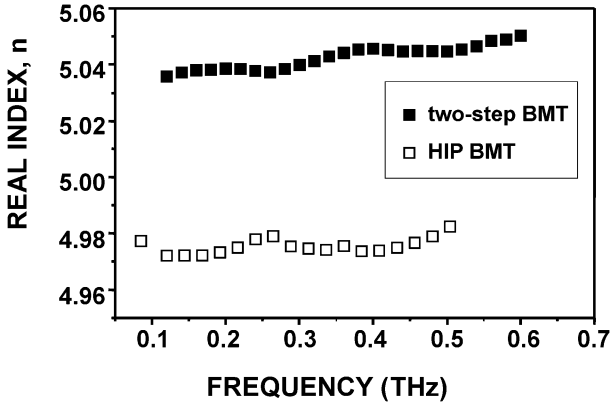

Fig. 4. The real index of refraction versus frequency for BMT (■, two-step processed BMT; □, hot isostatic pressed BMT).

In this case the relation between the complex index of refraction ($N_s = n + ik$) of sample and the transmission function can be written as

$$t(\omega) = \left(\frac{4N_s}{(N_s+1)^2}\right)\exp(i(k_s - k_v)d). \quad (1)$$

where $k_s = 2\pi\nu N_s/c$, $k_v = 2\pi\nu/c$, d is the thickness of the sample and c is the speed of light in vacuum. Eq. (1) can be solved numerically using a Newton–Raphson method. Part of the results obtained by numerical method can be found in Ref. 15. However, to circumvent the uncertainty possibly resulting from numerical process, we employed another, more precise method to solve the complex index of BMT. The complex transmission function for two samples of different thickness, which were sliced from the same piece of materials, was acquired. The two spectra, after Fourier transform, are divided to eliminate the effects of the uncollimated beam and the reflections from the sample surfaces. The real index of refraction n is obtained from the phase ϕ of the transmission function by

$$n = 1 + \frac{c(\phi_1 - \phi_2)}{2\pi\nu(d_1 - d_2)}. \quad (2)$$

where d_1 and d_2 are the sample thicknesses. The power absorption coefficient $\alpha = 4\pi\nu k/c$ is obtained from the amplitude A of the transmission function by

$$\alpha = \left[\frac{2}{(d_1 - d_2)}\right]\ln\left[\frac{A_2}{A_1}\right]. \quad (3)$$

The thicknesses of two-step BMT are 4.75, 2.44 and 2.40 mm and the thicknesses of HIP BMT are 5.10, 4.77 and 3.29 mm.

The results for the real index of BMT are shown in Fig. 4. In this figure, the value of the real index of two-step BMT extends from 5.03 to 5.05 in the 0.1–0.6 THz region. The real index of HIP BMT extends from 4.97 to 4.98. It is clear that the real index of HIP BMT is smaller than two-step BMT. Fig. 5 shows the power absorption α versus frequency curve. In this figure, both of the power absorption coefficient curves increase with increasing frequency. HIP BMT exhibits much larger power loss than two-step BMT. The value of the power absorption coefficient of two-step BMT extends from 0.07 mm^{-1} to 0.8 mm^{-1} in the 0.1–0.6 THz region. The values we obtain are consistent with earlier data.[15] The value of the power absorption coefficient of HIP BMT extends from 0.24 mm^{-1} to 1.20 mm^{-1} in the 0.1–0.5 THz region.

The Q-factor of BMT in the 0.1–0.6 THz region can be extracted from the real index n and the power absorption α and is shown in Fig. 6. It is clear that two-step BMT exhibit much higher Q than HIP BMT in this frequency range. The value of Q-factor of two-step BMT decreases with increasing frequency and varies from ~330 at 0.1 THz to ~77 at 0.6 THz. The Q-factor versus frequency curve of HIP BMT is much flatter in 0.1–0.5 THz region. Fig. 7 shows the Qf versus frequency curves. The $Q \times f$ of two-step BMT at 0.1 THz is ~40,000 GHz, which is slightly smaller than the $Q \times f$-values of two-step BMT materials in the microwave

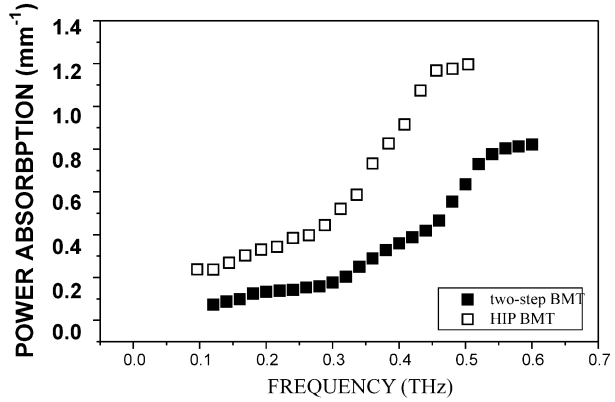

Fig. 5. Power absorption versus frequency for BMT (■, two-step processed BMT; □, hot isostatic pressed BMT).

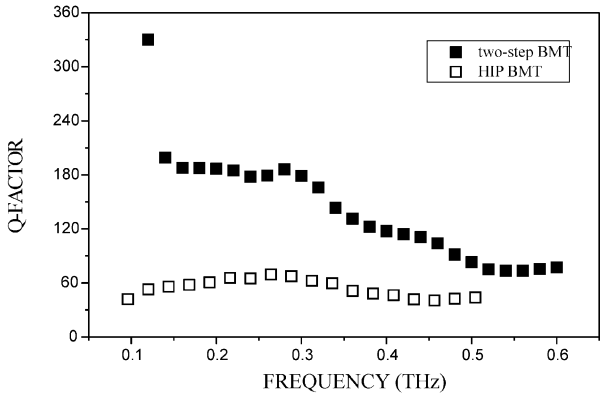

Fig. 6. The Q-factor versus frequency for BMT (■, two-step processed BMT; □, hot isostatic pressed BMT).

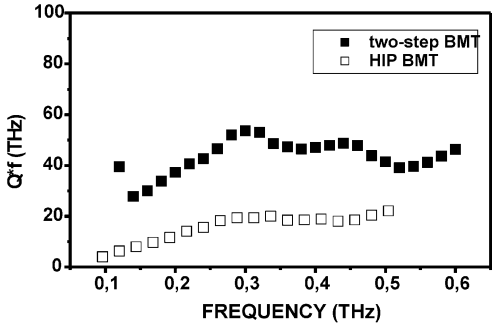

Fig. 7. The $Q \times f$ versus frequency for BMT (■, two-step processed BMT; □, hot isostatic pressed BMT).

regime ($Q \times f = 60{,}000$ GHz in 10 GHz). The $Q \times f$ of HIP BMT at 0.1 THz is only ~4,000 GHz.

It was proposed that the higher losses in some of the samples are related to the occurrence of the second phase, which exhibits a phonon peak at ~1.3 THz.[13] The submillimeter wave power absorption coefficient of BMT containing the second phase is about two times higher than the power absorption coefficient expected for the pure phase BMT.[12] Therefore, we can attribute that the higher power loss for HIP BMT is the existence of the second phase, although the 1.3 THz phonon peak is not clearly observed in our results due to the frequency range limit in our setup.

4. Conclusions

We used pulsed terahertz radiation to study the dielectric properties of BMT, which were prepared via a two-step process and a hot isostatic press process. The transmission spectrum of BMT in the 0.1–0.6 THz region was obtained. The real part of the index of refraction of two-step BMT in this band was roughly 5.04. The real part of the index of refraction of HIP BMT in this band was roughly 4.97, which is slightly smaller than the real part of the index of BMT ceramics in the microwave regime ($n = 5.0$). HIP BMT exhibit much higher power loss and lower Q-factor than two-step BMT. We can conclude that two-step process is a better way to prepare BMT than hot isostatic press process.

Acknowledgements

We acknowledge the financial support from the National Council of Science of ROC under grant No. NSC89-2622-E-007-005.

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