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Optical tuning of dielectric properties of YIG ceramics in the terahertz range

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Abstract: The tunability of the terahertz dielectric spectra of $Y_3Fe_5O_{12}(YIG)$ ceramics under external optical field were investigated at room temperature by using terahertz time-domain spectroscopy. The $Y_3Fe_5O_{12}$ ceramics annealed at 1 100 and 1 300 °C were successfully synthesized by a modified Pechini process. The samples were characterized by X-ray diffraction and no impurity phase has been detected. The crystallite sizes of the YIG ceramics are calculated to be about 50 nm for $T_s=1$ 100 °C and 77 nm for $T_s=1$ 300 °C, respectively. And Scanning Electron Microscope(SEM) images show that the grain size is positively correlated with the sintering temperature. Application of the optical field leads to an appreciable tuning of the dielectric constant of the sample sintered at 1 100 °C, which reaches up to 17.3% at 0.6 THz. Meanwhile the dielectric loss tangent of the sample sintered at 1 100 °C changes about 123.9%. On the contrast, both the dielectric constant and the loss tangent for the sample sintered at 1 300 °C keep invariable with the light excitation. These findings are attributed to space charges hopping among the grain boundaries excited by the external optical field.

Keywords: yttrium iron garnet; terahertz time-domain spectroscopy; dielectric property; space chargeCLC number: TN304Document code: Adoi: 10.11805/TKYDA2019340

1 Introduction

Yttrium iron garnet, $Y_3Fe_5O_{12}(YIG)$, a ferrite, has been widely applied in electronic devices for the microwave region, such as circulators, oscillators, phase shifters, isolators, and bubble-storage memory units. Because it possesses low dielectric loss and narrow resonance line width in the microwave region^[1], YIG has been attracting much interest in view of its dielectric and magnetic properties^[1-3]. A number of studies on the dielectric properties of YIG in wide temperature and frequency ranges have been carried out since the original work of Larsen and Metselaar^[4-5]. However, most investigations were conducted on single crystals of YIG^[6-8] or limited to frequencies below Ka-band(26-40 GHz)^[9]. Only a few studies focused on dielectric behaviors of polycrystalline YIG or the dielectric resonance beyond 40 GHz. M. A. Popov etc. have reported magnetically tunable dielectric resonances in YIG of single crystals at 40–110 GHz, its dielectric loss tangent is about 2×10^{-4} , and the dielectric properties of YIG can be tuned by the applied electric field^[10] and magnetic field^[11], which are promising candidates for tunable multifunctional devices. However, the dielectric properties with light excitation of polycrystalline YIG in the terahertz range have rarely been reported so far. As the rapid developments of terahertz time-domain spectroscopy(THz-TDS) and the spectroscopic applications of THz radiation in the last two decades^[12-15], it is necessary to study the dielectric responses of YIG at THz band to expand the application scope of YIG at higher frequencies.

In this paper, the single-phase polycrystalline YIG ceramics with different sintering temperatures ($T_s=1\ 100\ ^\circ$ C, and 1 300 $^\circ$ C) were prepared by a modified Pechini method. The transmittance spectra in the terahertz range (0.2–1 THz) were investigated under an external optical field and the dielectric properties of the YIG ceramics in the terahertz range were observed to be modulated under the external optical field at room temperature.

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2 Experiment

YIG ceramics were prepared by a modified Pechini method. First, yttrium nitrate hexahydrate $Y(NO_3)_3 \cdot 6H_2O$ (aladdin 99.9%) and iron nitrate enneahydrate $Fe(NO_3)_3 \cdot 9H_2O$ (aladdin 99.99%) were dissolved into a citric acid solution with glycol in stoichiometric ratio of Y:Fe=3:5. Citric acid (C₆H₈O₇•H₂O) (aladdin 99.8%) was added in 1:1.5 molar ratio with respect to the metal nitrates to aqueous solution. The pH value of the solution was adjusted to 7 by Ammonia. The precursor solution was evaporated and baked to the xerogel. The dried gel was ground to powder and heated at 600 °C in air for 6 h, and then pressed into pellets. Finally, the pellets were sintered to ceramics at various sintering temperatures (*T*s=1 100 °C and 1 300 °C) in air for 4 h with 10 mm diameter and 1.2 mm thickness. The ceramic density is 3.13 g/cm³ and 3.20 g/cm³ for the sample with *T*s=1 100 °C and 1 300 °C respectively.

The crystalline structures of the samples were recorded on Shimadzu XRD-6100 by using Cu K_{α} radiation. The particle size and morphology of the YIG ceramics were observed by using a field emission scanning electron microscopy

(SEM) (ZEISS, GeminiSEM 300, Germany). The transmittance spectra of the YIG ceramics were measured by a THz time-domain spectrum(THz-TDS) system (Zomega Terahertz Co., America). The frequency resolution was 10 GHz, the spot size at the focus was 3 mm, and the detectable frequency was ranged from 0.2 THz to 1 THz (300–1 500 μ m), including the atmospheric windows around 0.21,0.35, 0.41, 0.68,0.85,0.93 THz. An all-solid-state green laser(center wavelength of 532 nm) was employed as the external optical field. The laser was obliquely incident on the surface of the samples at an angle of 45° with a 5 mm diameter of spot size. All the measurements were obtained at room temperature(300 K) in a dry air environment.

3 Result and discussion

Fig.1 displays the XRD patterns at room temperature of YIG ($T_s=1\ 100\ ^{\circ}$ C and 1 300 $^{\circ}$ C). The cubic garnet structure can be easily observed in both the two samples, the crystalline phases are identified by using the JCPDS(Joint Committee on Powder Diffraction Standards) Powder Diffraction File (Card Nos.:70–0953), and no impurity phase is detected. According to the Scherrer equation, the crystallite sizes of the YIG ceramics are 50 nm for $T_s=1\ 100\ ^{\circ}$ C and 77 nm for $T_s=1\ 300\ ^{\circ}$ C, respectively. This indicates that the crystallite size of the ceramics is positively correlated with T_s .

The typical SEM images in Fig.2 of the YIG samples reveal that the distribution of grain sizes are homogeneous, and no segregation of impurity phase is detected. The grain sizes vary in the range of $0.5-1.2 \ \mu\text{m}$ and $1-1.7 \ \mu\text{m}$ for $T_s=1\ 100 \ ^{\circ}\text{C}$ and $T_s=1\ 300 \ ^{\circ}\text{C}$ respectively. This indicates that the grain size of the ceramics is positively correlated with T_s .



Fig.2 SEM images of the cross-sectional surface of YIG ceramics sintered at various temperatures (a) with $T_s=1~100$ and (b) 1300 °C

Fig.3(a) shows typical experimental results of the terahertz time domain waveforms passing through the YIG ceramics with $T_s=1\,100\,^{\circ}$ C and 1 300 $^{\circ}$ C. The time lag between the reference (air) and both the two samples is around 5.30 ps. By applying an external optical field of 200 mW, the time shifts are observed in the time domain. The transmission waveforms shift left by approximately 0.77 ps for $T_s=1\,100\,^{\circ}$ C and 0.06 ps for $T_s=1\,300\,^{\circ}$ C respectively when the light intensity is 200 mW, compared with that without the external optical field, as shown in Fig.3(b),(c).

Through a Fourier transform, we get the complex transmittances and the intrinsic phases in the frequency domain from the waveforms in Fig.3. The frequency-dependent refractive indexes, absorption coefficients and dielectric functions of the samples under external optical fields of 0 and 200 mW are calculated by using equations $(1)-(3)^{[16-19]}$:

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$$n = 1 + \frac{c}{\omega d}\phi \tag{1}$$

$$\alpha = -\frac{2}{d} \ln \left[\frac{\left| E_2(\omega) \right|}{\left| E_1(\omega) \right|} \frac{(n+1)^2}{4n} \right] = \frac{2\kappa\omega}{c} \tag{2}$$

 $\varepsilon = \varepsilon_{\rm r} + i\varepsilon_{\rm i} = (n + i\kappa)^2$ (3)

where $E_2(\omega)$ is the frequency waveform of the samples and $E_1(\omega)$ is the frequency waveform of the air, c is the speed of light, d is the thickness of the sample, α is absorption coefficient, κ is extinction coefficient, and n is the refractive index, ϕ is the phase of the complex transmission, ε is the complex permittivity, ε_r is the real part of ε , ε_i is the imaginary part of ε . Here we assume the magnetic susceptibility $\mu \approx 1$, since the contribution of magnetic susceptibility of YIG to the complex refractive index is much smaller compared to the dielectric one, even at the magnetic resonance frequency^[9,20].

Fig.4 shows the absorption coefficients and refractive indexes of the YIG ceramics with $T_s=1\,100$ °C and 1 300 °C under external optical field with the light intensity of 0 and 200 mW. In the entire frequency range of 0.2 THz to 1 THz, the absorption coefficient raises after applying light excitation for the sample sintered at 1 100 °C. At 0.6 THz, the absorption coefficient increases from 1.59 to 3.23. While for the sample sintered at 1 300 °C, the absorption coefficient shows unnoticeable change with light excitation. Meanwhile, the refractive index decreases as the external optical field is



applied to the sample sintered at 1 100 °C, for example, at 0.6 THz, the refractive index reduces from 1.89 to 1.72, while for the sample sintered at 1 300 °C, the refractive index shows no appreciable change with light excitation as well.



The dielectric properties of YIG in the terahertz range can be obtained by the refractive index and absorption coefficient according to Equation (3). As shown in Fig.5, it represents frequency dependent dielectric properties of the YIG ceramics with $T_s=1\,100\,^{\circ}$ C and 1 300 $^{\circ}$ C under external optical fields of 0 and 200 mW. The dielectric loss tangents of YIG ceramics with $T_s=1\,100\,^{\circ}$ C and 1 300 $^{\circ}$ C are lower than 3×10^{-2} at the whole frequency range. Besides, the permittivity decreases as the light excitation is applied over the whole measurement frequency range to the sample sintered at 1100 $^{\circ}$ C, while the loss tangent increases considerably with the light excitation. At 0.6 THz, the permittivity significantly decreases from 3.59 to 2.97 by 17.3%. The loss tangent increases from 0.006 7 to 0.015 by 123.9%. On the contrast, both the permittivity and loss tangent for the sample sintered at 1 300 $^{\circ}$ C keep invariable with light excitation.



Fig.5 Frequency dependence of (a)(b) real part permittivities and (c)(d) loss tangents of YIG ceramics (T_s =1100 and 1 300 °C) under external optical fields of 0 and 200 mW

In brief, for the YIG ceramic sintered at 1 100 °C, the refractive index and the permittivity decrease, yet the absorption coefficient and dielectric loss tangent increase after applying the external optical field with the light intensity of 200 mW. It is reported that at a high frequency range(above 10 kHz), the dielectric properties are highly dependent on the inhomogeneous structure, such as grain boundaries^[4]. The calculated activation energy of YIG is 0.83 eV for the grain boundaries. Therefore, in the illuminated area, with the laser beam of the wavelength of 532 nm, the space charges on the grain boundaries may hop to the conduction band, they migrate in a preferred direction to the dark area due to the photovoltaic effect, and are subsequently captured by other grain boundaries^[21]. The migration process results in an enhancement of the conductivity in the sample, which will then cause the increase of loss tangent tan δ . Meanwhile, the decrease of the permittivity can be attributed to the discharging of the space charges due to charge hopping exited by external optical field. By contrast, for the YIG ceramic sintered at 1300 °C, all the parameters show no considerable variation with the light excitation. It is due to the larger grains for the YIG ceramic sintered at 1 300 °C, there are no enough grain boundaries to generate light exciting charge carriers.

4 Conclusion

In summary, single-phase polycrystalline YIG ceramics were prepared by a modified Pechini method with $T_s=1\,100$ °C and 1 300 °C. The XRD patterns suggest that the grain size of the ceramics is positively correlated with T_s . We have investigated the frequency dependence of dielectric spectra of YIG ceramics sintered at 1 100 and 1 300°C and their optical-field-induced tunability by using terahertz time-domain spectroscopy at room temperature. The dielectric loss tangent is lower than 3×10^{-2} at the whole frequency range for both the two samples. An appreciable variation of dielectric

spectra in YIG ceramics sintered at 1 100 $^{\circ}$ C was also demonstrated with the external optical field. This property originates from the space charges hopping among the grain boundaries. Such a variation has not been detected in the sample sintered at 1 300 $^{\circ}$ C, this may attribute to the increment of crystallite size as the increase in the sintering temperature. The results will be helpful for the applications of YIG crystal in the THz field.

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