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Terahertz time domain spectroscopy of GdBCO superconducting thin films

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Abstract

We present terahertz optical properties of $GdBa_2Cu_3O_{7-x}$ (GdBCO) superconducting thin films. GdBCO films with a thickness of about 105 nm were grown on a $LaAlO_3$ (LAO) single crystal substrate using a conventional pulsed laser deposition (PLD) technique. Using an Ar ion milling system, the thickness of the GdBCO film was reduced to 58 nm, and its surface was also smoothened. Terahertz (THz) transmission spectra through two different GdBCO films are measured over the range between 0.2 and 1.5 THz using THz time domain spectroscopy. Interestingly, the THz transmission of the thinner GdBCO film has been increased to six times larger than that of the thicker one, while the thinner film is still maintaining its superconducting property at below 90 K.

Keywords: terahertz time domain spectroscopy, GdBCO, Ar-ion milling, Pulsed laser deposition

1. INTRODUCTION

Superconductors are fascinating materials, which show zero resistivities when they are cooled down below the critical temperature. Even though superconductors were discovered a century ago, they are recently opening up great possibilities for photonics and metamaterials as low-loss plasmonic devices operating at higher frequencies [1-6]. Metals have been used in many plasmonic and metamaterial structures for a few decades [7-9]. Although the patterned metals worked well in the microwave and lower frequency range, their ohmic loss gives rise to a lower quality factor (Q-factor) as their resonance frequencies increase to terahertz, infrared, and visible regions [10, 11]. Furthermore, it is almost impossible to actively control the intrinsic conductivity of a metal. Superconductors are great substitutes for metals at higher frequencies and maintain their minimal ohmic loss at below a critical temperature T_c. It should be noted that superconductors can be therefore exploited in metamaterials and nanogap structures operating at terahertz or lower frequencies, where THz electromagnetic waves would not easily break Cooper pairs [2, 6, 11-13].

Here, we report THz optical properties of GdBCO, one of high T_c superconductors, where its film thickness is as thin as about 50 nm. In order to measure a proper THz transmission signal through the superconducting film at below T_c, the film thickness should be about 100 nm or below. We prepared a 105 nm thick GdBCO film on $LaAlO_3$ (LAO) substrate using conventionalPLD technique. To further reduce the film thickness, an Ar-ion based milling system is used in this experiment. To optically characterize the superconducting thin films in the THz region, we performed a transmission-type THz time domain spectroscopy with a low temperature cryostat (Fig. 1). The ion-milled 58 nm-thick GdBCO film have proved to exhibit excellent THz transmission characteristics, compared to the 105 nm-thick GdBCO film.

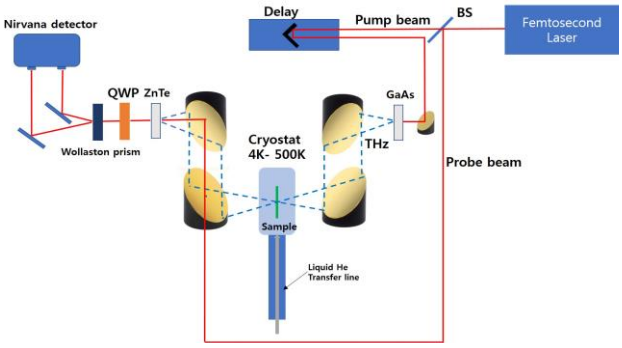

Fig. 1. Schematic diagram of a transmission-type THz time domain spectroscopy based on a GaAs photoconductive antenna and a 1 mm-thick ZnTe (110) crystal as a detector with a low temperature cryostat. QWP: Quarter Wave-Plate. BS: Beam Splitter.

2. EXPERIMENTAL METHODS

2.1. THz time domain spectroscopy

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To experimentally probe the THz response of our superconducting thin films, we performed THz time domain spectroscopy (0.2 ~ 2.5 THz) [14, 15] with a single-cycle picosecond pulse, which is generated from a commercial GaAs emitter (Tera-SED, Laser Quantum) illuminated by a Ti:sapphire laser pulse train with 800 nm center wavelength, 75 MHz repetition rate, and sub-10 fs pulse width (Synergy, Spectra-Physics), as shown in Fig. 1. To strictly control the temperature of the sample, we put a low temperature cryostat (4~500 K) on a focal point of the THz beam. To choose a proper substrate for the GdBCO film, we measured the transmission spectra of two 500 μm-thick substrates of $LaAlO_3$ (LAO) and $SrTiO_3$ (STO), which are typically used for growth of GdBCO films. Fig. 2(a) shows the measured electro-optic signals through the bare aperture, a LAO substrate, and a STO substrate. The reference signal through the bare aperture is shown as the black dashed line. Fourier-transforming the time traces and dividing them by the reference signal, we get normalized transmitted amplitude spectra in Fig. 2(b). As shown in Fig. 2(b), it is evident that the LAO substrate is much more transparent to THz electromagnetic waves compared to the STO substrate.

2.2. Sample fabrications

GdBCO films with a thickness of about 100 nm were fabricated on $LaAlO_3$ (LAO) single crystal substrates by using a conventional PLD technique. A KrF excimer laser with a wavelength of 248 nm operated at an energy of 200 mJ and the repetition rate of 3 Hz was focused on a GdBCO target, prepared by the solid state reaction method. The substrate temperature and oxygen gas pressure were kept at 750 °C and 200 mTorr, respectively. After completing depositions, all of the films were annealed at 500 °C in 500 Torr oxygen for one hour and cooled down to room temperature. The superconducting transition temperature was determined by measuring resistance as a function of temperature by using a standard four probe technique in a closed cycle refrigerator. The thickness of the completed GdBCO film is 105 nm, as confirmed by the SEM image in Fig. 3(a).

We performed the ion milling process to reduce the thickness of the GdBCO superconducting thin films. We used Ar-ion miller (KVET-IM2000L, Korea Vacuum Tech) with 80 V acceleration voltage and 1.7 mA beam current. We reduced the 105-nm-thick GdBCO thin film to 58 nm with an Ar ion beam at an oblique angle of 75 ° and an exposure time of 10 minutes. After ion-milling, the actual thickness of the GdBCO film becomes 58 nm, and the surface roughness is relatively smoothened, as shown in Fig. 3(b).

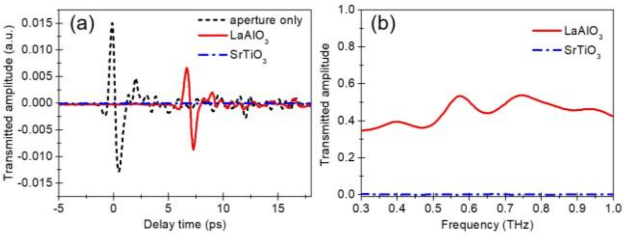

Fig. 2. (a) Electro-optic sampling signal in time-domain, through the bare aperture (black dashed), LAO (red solid), and STO (blue dashed dot). (b) Transmitted electric field amplitude spectra of LAO and STO.

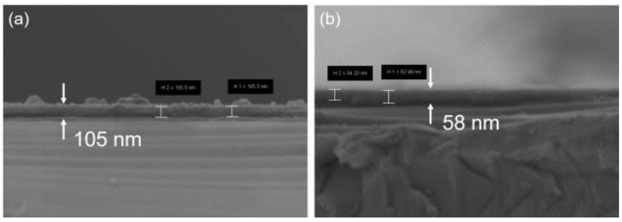

Fig. 3. Scanning electron microscope (SEM) images of the cross-section of the two GdBCO films with the thicknesses of (a) 105 nm and (b) 58 nm.

3. RESULTS AND DISCUSSIONS

Fig. 4(a) compares THz time-domain signals transmitted through the 58 nm-thick GdBCO film depending on the temperature in the range between 22 K (black) and 200 K (red). Thanks to the small thickness of the GdBCO film, the THz transmission through the superconducting film remains high enough to be measured even at below its transition temperature of about 90 K. After Fourier-transforming the THz time traces in Fig. 4(a), we get the THz transmittance spectra in the frequency range between 0.2 and 1.5 THz in Fig. 4(b). It shows that the low frequency transmittance quickly decreases as the superconductivity develops.

We now discuss temperature-dependent transmittance curves at a specific frequency of 0.7 THz for the two different GdBCO films with the thicknesses of 105 and 58 nm. In Fig. 5, a linear decrease of the transmittance has been observed with the decreasing temperature to above transition temperature of 90 K. However, abrupt changes in the transmittance of both samples are seen at around 90 K in terms of exponential decrease of the transmittance curve towards the lower temperature. This change reflects a superconducting property of the GdBCO thin films at below T_c, even after the ion-milling process. It should be also noted that the THz transmittance through the 58 nm-thick GdBCO film has been increased to 0.063, 6 times

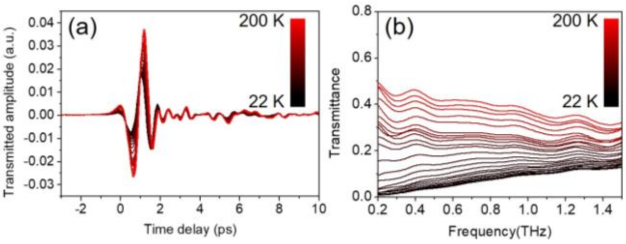

Fig. 4. (a) THz time-domain signals through the 58 nm-thick GdBCO film depending on the temperature in the range of 22 K and 200 K. (b) Fourier-transformed THz transmittance spectra in the frequency range between 0.2 and 1.5 THz.

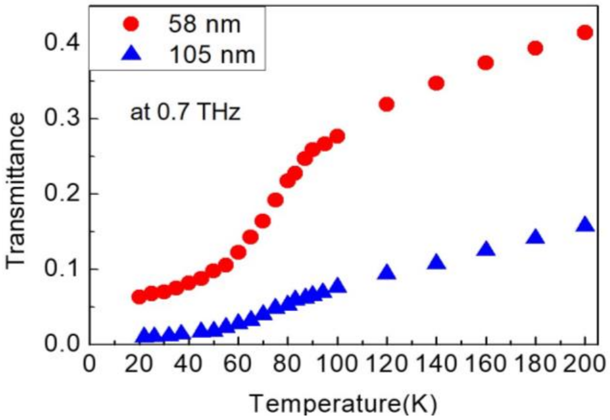

Fig. 5. Temperature-dependent transmittance curves of the two GdBCO films with the thicknesses of 105 nm (blue triangle) and 58 nm (red circle) at 0.7 THz.

larger than 0.0097 of the 105 nm-thick GdBCO film at 22 K. This means that the transmittance through the 58 nm-thick superconducting film would be clearly measured.

In conclusion, we experimentally demonstrate THz transmission properties of the two GdBCO films with the thicknesses of 105 and 58 nm grown on the LAO substrate. Ar-ion milling works well in thinning down to obtain 50 nm-thick films, still maintaining their superconducting property at below T_c. Those thinner superconducting films are suitable to develop various THz applications combined with plasmonic nanostructures to enhance light-matter interactions [15-20]. Furthermore, this combined system would create opportunities in studying electrodynamics of Cooper pairs inside the limited volume underneath the plasmonic structures.

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