Terahertz Radiation Properties of Underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ Thin Films

Y. Tominari,$^{1,2}$ H. Murakami,$^{1,*}$ and M. Tonouchi$^{1,2}$

$^1$Research Center for Superconductor Photonics, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871 Japan

$^2$CREST, Japan Science and Technology Corporation (JST), 2-1 Yamada-oka, Suita, Osaka 565-0871 Japan

Terahertz (THz)-wave pulse radiation properties by femtosecond optical pulse excitation were investigated on high-$T_c$ superconductors. For the measurements three kinds of underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thin films with superconducting transition temperatures of $T_c = 84.0$ K, 61.5 K and 52.4 K were prepared by annealing with a bulk YBCO sample under different reduced oxygen conditions. It was found that the amplitude of the radiated THz pulse strongly depends on the hole-doping level. The maximum radiation efficiency was obtained for a middle underdoped sample with $T_c = 61.5$ K. The observed hole-doping dependence of radiation efficiency is qualitatively explained by the relation between the transmittance of YBCO and supercarrier density.

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I. INTRODUCTION

Generation technique of terahertz (THz)-wave pulse from semiconductor photoconductive switches (Auston switch) was developed by Auston et al. in 1983 [1]. Since then, much progress has been made in this research field by the discovery of various THz radiation sources such as semiconductors, manganite and superconductors [2–4]. THz-wave is located in the boundary region between microwave and infrared, which is wavelength of 3 mm~300 $\mu$m, frequency of 100 GHz~10 THz and energy 0.41 meV~41.3 meV. This region is so called undeveloped electromagnetic wave region which is expected to be developed for next generation microwave photonics. THz radiation mechanism from semiconductors is qualitatively explained by ultrafast excitation of photo-conductive carriers and local polarization induced by femtosecond optical pulse excitation. Since the ultrafast modulation process takes place in a time-scale of several subpicoseconds, THz pulse can be radiated from semiconductors.

On the other hand, current biased high-$T_c$ superconductors (HTSC) also radiate THz-wave pulse due to ultrafast supercurrent modulation induced by femtosecond optical pulse excitation [5]. We have up to now investigated THz radiation properties of various kinds of HTSC materials [6, 7], and demonstrated that the radiation property is deeply related to the intrinsic physical properties of each sample. Particularly, the amplitude and frequency components of the THz pulse are considerably different from sample to sample.

In the present study, to investigate the origin of the difference in the THz-wave pulse radiation properties of HTSC, THz-wave pulse radiation was observed on systematically
underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) samples with different hole-doping levels and critical temperatures $T_c$.

II. EXPERIMENTAL DETAILS

For the measurements we prepared $c$-axis oriented YBCO thin films on MgO substrate ($10 \times 10 \times 0.5$ mm$^3$). The typical thickness of the film is 200 nm. The hole-doping level of the film was controlled by annealing under the various oxygen partial pressures, $P_{O_2}$, with an YBCO polycrystalline pellet. A $12 \times 12 \times 0.5$ mm$^3$ square hole was formed in the surface of the pellet to set the film sample with an MgO substrate, and the surface of the thin film was placed on the pellet in contact with the bottom of the hole. A set of the pellet and thin film was annealed in a furnace at 600$^\circ$C for about 24 h under the various oxygen partial pressure conditions of $P_{O_2} = 1$ atm (sample-A), $P_{O_2} = 0.5$ atm (sample-B) and $P_{O_2} = 0.1$ atm (sample-C). After the annealing process, the films were quenched at room temperature. Characterization of the annealed samples was carried out by X-ray diffraction measurement. To investigate the THz-wave pulse radiation properties, the films were patterned into $20 \times 20 \mu$m$^2$ (width×length) dipole antenna structure by means of a conventional photolithography and ion milling process. After patterning the film, temperature dependence of resistance along the bridge was measured by four probe method, and superconducting, $T_c$, and critical current density, $J_c$, were determined.

On the other hand, Fig. 1 shows a THz-wave pulse generation and detection system. A mode-locked Ti:sapphire laser operating at repetition rate of 82 MHz was used to generate the femtosecond laser pulses with a wavelength of 800 nm. The optical pulse is separated into pump [for sample excitation] and trigger pulse [for detector]. The pump pulse was focused onto the current biased dipole antenna at a spot size of 30 m in diameter.
Fig. 2 and the inset show the temperature, $T$, dependence of the resistance along the bridge, $R$, and critical current density, $J_c$, for prepared YBCO dipole antennas, respectively. It can be seen that the value of $J_c$ decreases from sample-A to -C with decreasing the oxygen partial pressure for annealing process. From the $R$-$T$ measurements, the critical temperature, $T_c$, was determined as $T_c = 84.0$ K for sample-A, 61.5 K for -B and 52.4 K for -C. On the other hand, the X-ray diffraction patterns showed only (00$l$) peaks reflecting c-axis orientation for these samples. From the X-ray diffraction measurements c-axis lattice constants were determined as 11.702 Å (sample-A), 11.730 Å (sample-B) and 11.733 Å (sample-C). The obtained relation between $T_c$ and the c-axis parameter shows a good consistence with previous data. Using the data obtained by J. D. Jorgensen et al. and W. Prusseit et al. [9, 10], the $\delta$ value of YBCO can be estimated as $\delta \sim 0.21$ for sample-A, $\delta \sim 0.42$ for sample-B and $\delta \sim 0.46$ for sample-C, respectively.

Fig. 3 shows typical time-domain waveforms of THz-wave pulse radiated from sample-A to -C, which were observed under the same condition at pump power of 10 mW, bias current of 40 mA and temperature of 22 K. The pulse width of emitted THz-wave pulse expands gradually from sample-A to -C. The bias current dependence of maximum amplitude of the positive pulse of sample-A is shown in Fig. 4. The maximum amplitude increases almost linearly with increasing the bias current in the region below 100 mA, and more rapidly
FIG. 3: Typical time-resolved waveforms of the THz-wave pulse radiated from YBCO dipole antenna at 23 K. The measurement was performed at a pump power of 10 mW and bias current of 40 mA.

FIG. 4: Maximum signal amplitude near 15 ps in the THz-wave pulse of sample-A as a function of the bias current dependence at 22 K. The inset is pump power dependence with $I = 10$ mA and $T = 22$ K.

increases above 100 mA. This phenomenon could be explained by taking vortex flow phenomenon into consideration. In the THz-wave radiation from HTSC, electromagnetic wave is radiated by ultrafast modulation of supercurrent induced by femtosecond pump pulse excitation according to the following relation [5].

$$E = \frac{\partial J_n}{\partial t} = q n_s \frac{\partial v}{\partial t} + q v \frac{\partial n_s}{\partial t} \propto J, P_{\text{pump}}. \quad (1)$$

Here, $n_s$ is the supercarrier density, $v$ the velocity of supercarrier, and $P_{\text{pump}}$ the power of pump pulse. Namely, the amplitude of the radiated THz-wave pulse is almost proportional to the applied bias current and excitation pump power. However, the Lorentz force between
the applied bias current and the magnetic fluxes produced by the current gets strong with increasing the bias current. When the excitation pump pulse is illuminated to the bridge under this circumstance, it is easily supposed that vortex flow occurs. Since the vortex flow introduce a ultrafast transition from superconducting state to normal state, substantial enhancement in the radiation efficiency of THz-wave should take place. Because the transmittance of THz-wave pulse through YBCO in normal state is much higher than that in superconducting state. On the other hand, the inset shows pump power dependence of maximum peak amplitude of sample-A. It can be seen that the amplitude increases approximately linearly with the pump power in this region.

Fig. 5 simultaneously plots the bias current dependence of the peak amplitude for three samples with different hole-doping level. It shows the maximum amplitude for the sample-B with a middle hole-doping level over the whole regions. On the other hand, the lightly underdoped sample-A shows the lowest amplitude.

As for the obtained hole-doping dependence of the amplitude of the radiated THz-wave pulse, it reflects the close relation between the radiation efficiency of THz-wave and several physical factors, such as supercarrier density $n_s$, the transmittance for THz-wave, etc. It is considered that the transmittance of THz-wave through YBCO should increase with decreasing the hole-doping level, because the shielding effect against THz-wave (or dielectric constant, refractive index) gets weak (or small value) with reducing the hole carrier density in YBCO. On the contrary, the substantial intensity of generated THz-wave should decrease with reducing the hole carrier (supercarrier) density according to eq.(1). These relations give an opposite tendency in the radiation efficiency of THz-wave pulse against the hole-doping level. Therefore, the maximum or minimum point of the radiation efficiency naturally appears in the hole-doping dependence. These obtained results show that control of the hole-doping level is important to develop a HTSC THz-wave radiation source with high radiation efficiency.
IV. CONCLUSION

We observed THz radiation from underdoped YBCO thin films by femtosecond laser pulse excitation. The underdoped YBCO thin films were prepared by annealing with YBCO bulk pellet under the various reduced oxygen pressures. The physical parameters of the obtained samples show well regulated properties of underdoped YBCO. From the THz-wave pulse radiation studies, it was found that the radiation efficiency strongly depends on the hole-doping level, and the maximum efficiency was obtained for middle underdoped YBCO with $T_c = 61.5$ K.

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* Electronic address: murakami@rcsuper.osaka-u.ac.jp