Free-Space Electrooptic Sampling of Terahertz Radiation From Optically Excited Superconducting $YBa_2Cu_3O_{7-\delta}$ Thin Films

Po-Iem Lin, K. H. Wu, J. Y. Juang, J.-Y. Lin, T. M. Uen, and Y. S. Gou

***Abstract*—The characteristics of photogenerated terahertz radiation from a current-biased superconducting $YBa_2Cu_3O_{7-\delta}$ (YBCO) bow-tie antenna were investigated using a free-space electrooptic sampling technique. Picosecond electromagnetic pulses about 450 fs wide were obtained. The frequency spectrum derived by Fourier transforming the picosecond pulses spans over 0.1–4 THz. The dynamics of the quasi-particles optically induced by the ultrafast laser pulse primarily determines the performances of the transient terahertz radiation generated under different operating parameters. The results indicate a characteristic quasi-particle relaxation time of about 2.5 ps close to the critical temperature T_c, and a faster time at lower temperatures.**

***Index Terms*—Femtosecond laser illumination, free-space electrooptic sampling (FSEOS), nonequilibrium superconductivity, terahertz radiation, $YBa_2Cu_3O_{7-\delta}$ (YBCO).**

I. INTRODUCTION

THE ULTRAFAST photoresponse of high-T_c superconductors (HTSCs) has attracted much attention due to its unique capacity in uncovering the nonequilibrium dynamics of the optically excited quasi-particles. The optical reflectivity measured by the pump-probe method has demonstrated femtosecond time response in the superconducting state [1]–[4]. Using a subpicosecond electrooptic sampling system, the picosecond electrical response from a current-biased $YBa_2Cu_3O_{7-\delta}$ (YBCO) microbridge exposed to pulsed femtosecond optical radiation was observed [5], [6]. Recently, observation and detection of picosecond electromagnetic pulses emitted from optically excited superconducting bridges have further revived the interest of using HTSC films as potential terahertz radiation sources [7]–[10]. However, depending on the radiation and detection schemes employed, the resultant waveforms and frequency bandwidths were different and had been a subject of extensive research. For instance, Cai *et al.* [11] and Park *et al.* [12] have made a direct comparison between the performances and detected waveforms of free-space electrooptic sampling (FSEOS) and photoconductive antenna (PCA) detection schemes to detect coherent terahertz radiation emitted from biased low-temperature-grown GaAs emitters.

In this paper, we report the first observation of terahertz generation from superconducting YBCO thin films by using a FSEOS system. The polarization change of the probe beam occurs when the ZnTe sensor crystal, based on the Pockels effect, is irradiated by the terahertz electric field. The terahertz-induced phase retardation of the probe beam is converted into an intensity modulation and detected by using balanced photodiodes. Picosecond electromagnetic pulses about 450 fs wide were obtained. The representative frequency spectrum derived by Fourier transform spans over 0.1–4 THz. We investigated the performances of the transient terahertz radiation by measuring the dependence of the terahertz radiation on excitation power, bias current, and ambient temperature. The results show that the peak strength of the transient terahertz radiation increases linearly with optical power as well as the bias current, proving the superradiant character of the emission. At lower ambient temperature, the behaviors of the transient terahertz radiation are all the same indicated that there exists a similar characteristic time of the quasi-particle recombination. Toward the critical temperature T_c, a slower recovery component of characteristic time 2.5 ps of the transient terahertz waveforms related to the quasi-particle recombination process across a smaller energy gap was observed. The results imply that the dynamics of the emitted terahertz transient is closely related to nonequilibrium superconductivity in the YBCO.

II. EXPERIMENT

A. Sample Preparation

YBCO thin films were deposited on 0.5-mm-thick MgO(100) substrates by pulsed laser deposition. The films were c axis oriented and had a typical thickness of 100 nm. The YBCO thin films were patterned into a bow-tie antenna structure having a center bridge length of 200 and width of 100 μm with the bow-tie angle of 60° by using standard photolithography and wet chemical etching. The critical temperature T_c was 88 K after patterning into the antenna structure, and the critical current density J_c were 1.7×10^6 A/cm^2 at 77 K and 1.0×10^7 A/cm^2 at 50 K, respectively.

B. Optical Setup

The experimental setup for the generation and detection of terahertz radiation using a FSEOS system is illustrated schematically in Fig. 1. A continuous wave (CW) argon-laser-pumped,

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P.-I. Lin, K. H. Wu, J. Y. Juang, T. M. Uen, and Y. S. Gou are with the Department of Electrophysics, National Chiao Tung University, Hsinchu 30050, Taiwan, R.O.C. (e-mail: glinpi.ep87g@nctu.edu.tw).

J.-Y. Lin is with the Institute of Physics, National Chiao Tung University, Hsinchu 30050, Taiwan, R.O.C.

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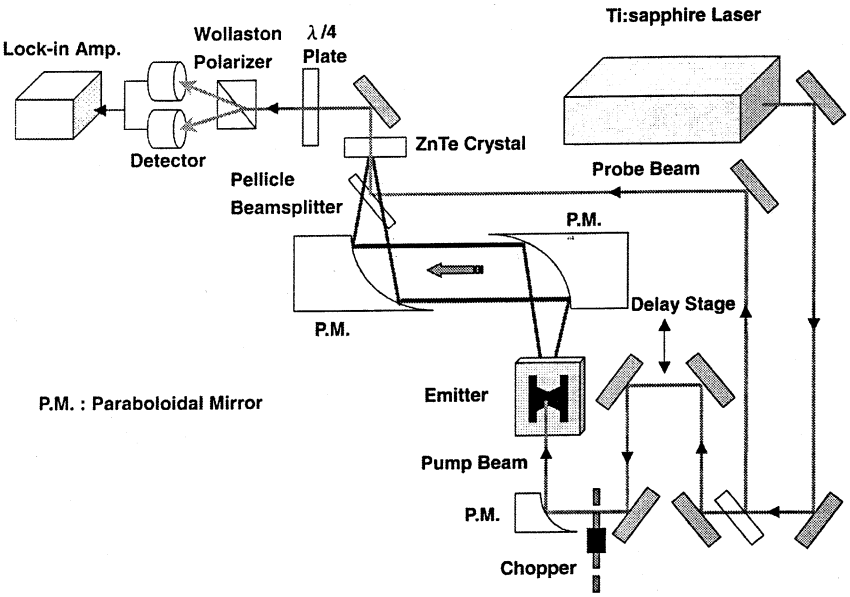

Fig. 1. Schematics of the experimental setup of FSEOS system.

compact mode-locked Ti : sapphire laser (Femtosource C20) provides 20-fs optical pulses at 800 nm (1.55 eV) with a 75-MHz repetition rate. The pump beam with spot size about 50 μm in diameter is modulated by a mechanical chopper which operated at 1.3 kHz and incident normal to the center bridge of YBCO bow-tie antenna. The electric field of the terahertz pulse is sampled by scanning the delay between the pump and probe beam. The superconducting YBCO bow-tie antenna, triggered by femtosecond optical laser pulses, radiates the terahertz beams. The terahertz radiation emitted from the backside of the MgO substrate is collimated by an MgO hemispherical lens with a diameter of 5 mm attached to the backside of the MgO substrate. Then, the terahertz radiation passed through a 3-mm-thick vacuum window made of Teflon and focused by a pair of off-axis paraboloidal mirrors onto the ZnTe sensor crystal. For low-temperature measurements, the samples were cooled using a Janis flow-through cold-finger cryostat. Further details of the experimental setup can be found in our previous publication [13].

C. Orientation Dependence of Terahertz Detection in ZnTe

Fig. 2 shows the measured maxima of the terahertz amplitude as a function of the azimuthal angle α when the probe beam polarization is parallel to the terahertz beam polarization. The orientation dependence of terahertz beam detection in ZnTe was accomplished by using an undoped semi-insulating GaAs (SI-GaAs) photoconductive switch [13]. Here, the angle α was defined as the angle of the terahertz beam polarization and the probe beam polarization with respect to the (001) axis of the (110)-oriented ZnTe crystal. These results show the optimal operating parameters for terahertz pulse detection using the ZnTe crystal [14].

III. RESULTS AND DISCUSSION

Fig. 3 shows the typical photogenerated terahertz radiation as a function of the scanning delay time obtained from a current-biased superconducting YBCO bow-tie antenna measured at 50 K. The excitation power and bias current (I_b) were 200 mW and 440 mA, respectively. A sharp pulse about 450 fs wide is observed. The polarity of the terahertz radiation waveform is opposite by reversing the bias current direction, and no signal is observed when the bias current is not applied. The polarization of the terahertz radiation is parallel to the bias current direction at the bridge. The modulation after the main pulse is measured under different operating parameters as will be discussed later. A representative power spectrum derived by Fourier transform of the terahertz waveforms is shown in the inset of Fig. 3. The radiation frequency spectrum extends from 0.1 to 4 THz with its peak intensity at 0.6 THz. The bandwidth at half-maximum (BWHM) of the frequency spectrum is around 1.1 THz. The corresponding atmospheric water vapor absorption at 1.1, 1.4, and 1.7 THz was observed.

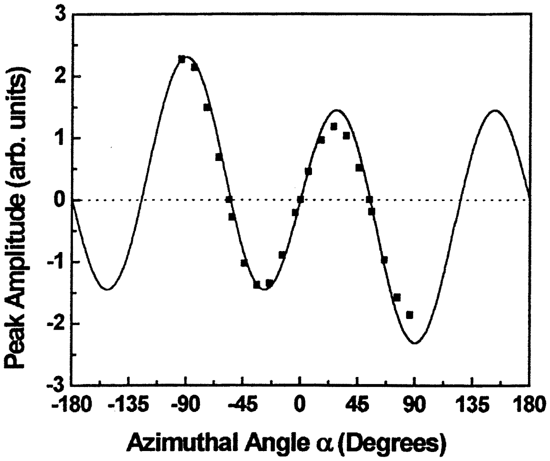

Fig. 2. Measured maxima of the terahertz amplitude as a function of the (110)-oriented ZnTe crystal's azimuthal angle α, where the terahertz measurements (square) and fit curve [14] (solid line).

Fig. 4 shows plots of a series of transient terahertz waveforms as a function of excitation power for the superconducting

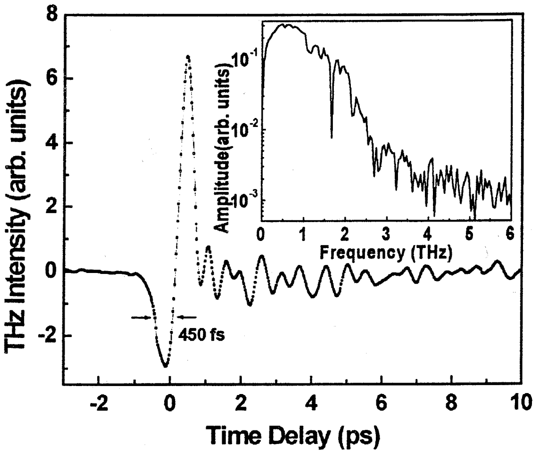

Fig. 3. Measured transient terahertz radiation from superconducting YBCO bow-tie antenna at 50 K. The excitation power and bias current were 200 mW and 440 mA, respectively. The inset shows the frequency spectrum by Fourier transform of the terahertz waveform.

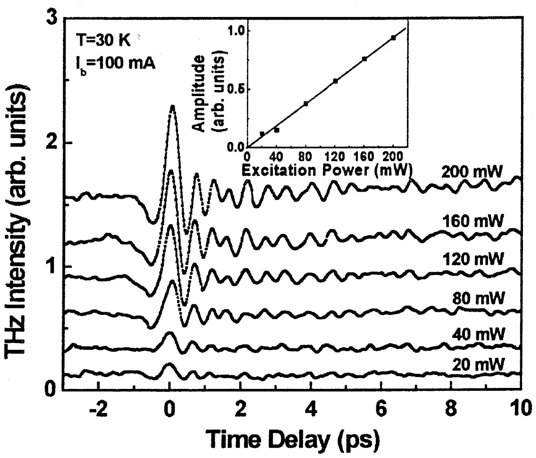

Fig. 4. Series of detected transient terahertz waveforms as a function of excitation power. The inset displays the linear relationship between the terahertz peak strength and the excitation power.

YBCO bow-tie antenna. The peak strength of emitted terahertz fields increases linearly with optical excitation power as shown in the inset of Fig. 4. It is evident that modulation after the main pulse is always present over the whole range of optical excitation power studied. Besides, the bias current dependence of the transient terahertz radiation is illustrated in Fig. 5. The peak amplitude increases in proportion to the bias current up to about 200 mA. Beyond this value, though it is still much less than the critical current I_c ($I_c = 650$ mA at 60 K), the peak amplitude increases rapidly due to the flux flow effect [15]. The resistive state resulted from laser heating or flux flow would produce a normal region that weakens the screening effect and absorption in the YBCO films and hence increases the emission efficiency of the terahertz radiation. The fact that peak amplitude is proportional to both the excitation power and the bias current, evidencing the superradiant character of the emission [9].

As far as classical electromagnetic dynamics is concerned, a far-field radiated terahertz electric field is proportional to the time derivative of the net current. From the previous results, it is natural to suggest that the terahertz radiation from YBCO films

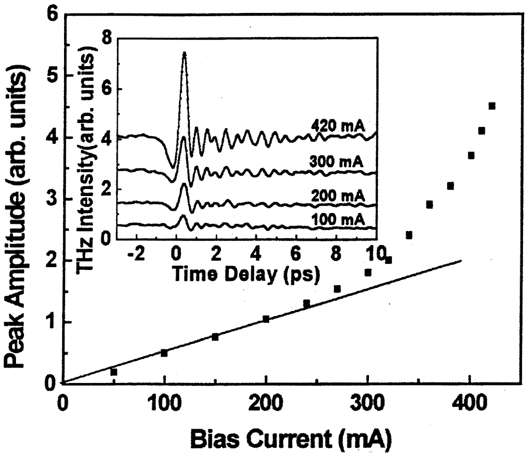

Fig. 5. Bias current dependance of detected terahertz peak amplitude. The solid line is drawn to indicate the trend. The inset shows the terahertz waveforms for various the bias currents. The excitation power is 120 mW measured at 60 K.

is generated by the modulation of the supercurrent. Namely, the transient waveform can be interpreted as the first derivative of the supercurrent in the films, and the time integral of the observed amplitude corresponds to the current transient in the time domain. The obtained terahertz radiation emitted from YBCO thin films by using the PCA detection technique were reported by Hangyo *et al.* [10]. The radiation mechanism was attributed to ultrafast supercurrent modulation by the laser pulses, which induce the nonequilibrium superconductivity. In that case, the nonequilibrium superconducting state is directly monitored as a transient change of the supercurrent. As can be seen, the decrease in the current is determined by multiple excitation of the quasi-particles due to hot-carrier thermalization. The decrease in the number of Cooper pairs results in the decrease in the supercurrent. The current recovery may be explained in terms of the recombination of quasi-particles into Cooper pairs. The previously mentioned characteristics are consistent with the results obtained here. Moreover, the bandwidth of the frequency spectrum we observed is wider and the modulation of the terahertz waveforms after the main pulse, detected by using FSEOS, is observed for all the excitation power and bias current applied. The appearance of the modulation of the terahertz radiation may be attributed to the high-frequency components on the superconducting YBCO thin films [8]. Detailed analysis is underway.

From the behaviors of bias current and excitation power, it is, thus, interesting to see whether the waveforms and frequency spectra would change with ambient temperature. Fig. 6 shows the temperature dependence of emitted terahertz waveforms. It is evident that the transient terahertz waveform behaviors are the same in each case, except in the results at 75 K, in which the waveform after the main pulse followed a slower component with a time of about 2.5 ps. Namely, the negative signals of the main pulse attributed to the decrease in the supercurrent have no significant temperature dependence, and the positive ones had different waveform behaviors that may be related to the quasi-particle recombination processes near the critical temperature T_c of the YBCO films (the increase in temperature will be about 10 K with the excitation power in our case). Besides, the

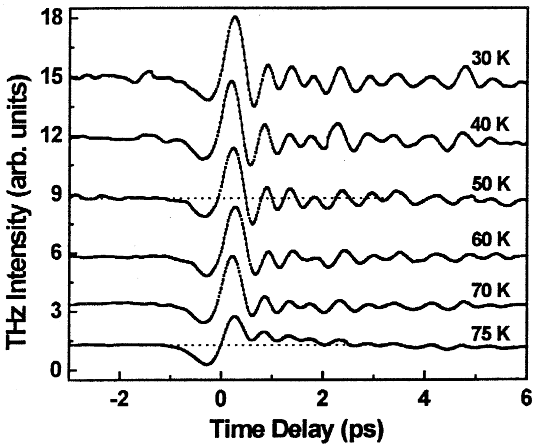

Fig. 6. Temperature dependance of emitted terahertz waveforms. The signal intensities were normalized by the negative peak strength of the main pulse of terahertz waveforms. The excitation power is 100 mW.

modulation after the main pulse is maintained by varying the ambient temperature as well as the excitation power and bias current. With regard to the peak amplitude of terahertz signals, the radiation amplitude rapidly increases with increasing ambient temperature and can be explained by the thermal shrinkage of the superconductor energy gap and of the temperature dependent transmission and absorption coefficients [9], [16].

Usually, the optical reflectivity measured by the optical pump-probe method has femtosecond time response while the gap opening is manifested by a rapid increase in the amplitude of the photogenerated transient reflectance in the superconducting state. The ultrafast rise of the reflectivity after excitation of the YBCO at $t = 0$ ps is attributed to Cooper pairs breaking and the subsequence decrease of the reflectivity results from quasi-particle recombination [1]–[3]. The recombination time, which has the value of 1.5–2.5 ps, as reported by Jaekel *et al.*, shows a crucial increase up to the value of 2.5 ps as the ambient temperature increases toward T_c. The divergent performance of the quasi-particle recombination is generally attributed to the manifestation of superconducting gap. Besides, Williams *et al.* demonstrated the superposition of the kinetic-inductive and the resistive hot-electron photoresponse mechanisms to explain the transformation of the voltage transient single-picosecond bipolar waveform to a broad photoresponse pulse at higher optical powers near T_c [5], [6]. In the present case, we interpret the emitted transient terahertz radiation as the ultrafast supercurrent modulation and found that the recovery time 2.5 ps of current near T_c is consistent with the result of Jaekel *et al.* [9]. In addition, the performance of the recovery processes is nearly the same below T_c indicated that there exists a similar characteristic time of the quasi-particle recombination. However, we found that the recombination time will vary with the operating parameters. At lower ambient temperature, the transient terahertz waveform behaviors are the same in each case. On the contrary, more excitation power results in a longer recombination time leading to lower peak intensity of emitted terahertz frequency spectrum as the ambient temperature nears the critical temperature T_c. That is to say, if one can offer appropriate excitation power and suitable ambient temperature, a series of transient terahertz radiation waveforms with different recombination times can be obtained.

IV. CONCLUSION

We report the first observation of terahertz generation and detection from current-biased superconducting YBCO thin films excited by femtosecond optical pulses by using a FSEOS technique. The picosecond electromagnetic pulses (450-fs width), generated by the terahertz-field-induced phase retardation of the probe beam converted into an intensity modulation, were obtained. The representative frequency spectrum derived by Fourier transform spans over 0.1–4 THz. The dynamics of the emitted terahertz transient related to the nonequilibrium superconductivity is investigated by measuring the dependence of the radiation on excitation power, bias current, and ambient temperature. The present results can be considered as a useful manner to probe the nonequilibrium superconductivity with the femtosecond time-resolved spectroscopy.

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