Photoresponse of YBa$_2$Cu$_3$O$_{7-\delta}$ Granular and Epitaxial Superconducting Thin Films

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ABSTRACT

In this paper we report on the response of thin films of YBa$_2$Cu$_3$O$_{7-\delta}$ with either a very grainy or a smooth epitaxial morphology to visible radiation. $\Sr$TiO$_3$ substrates were employed for both types of films. The grainy films were formed by sequential multi-layer electron beam evaporation while the epitaxial films were formed by laser ablation. Both films were patterned into "H" shaped detectors via a negative photolithographic process employing a Br/ethanol etchant. The bridge region of the "H" was 50$\mu$m wide. The patterned films formed by laser ablation and sequential evaporation had critical temperatures of 74 K and 72 K respectively. The bridge was current biased and illuminated with chopped He-Ne laser radiation and the voltage developed in response to the illumination was measured. A signal was detected only above the critical temperature and the peak of the response coincided with the resistive transition for both types of films although the correspondence was less exact for the grainy film. The details of the responses and their analysis are presented.

1. INTRODUCTION

The discovery of high temperature superconductors has prompted a large amount of research into potential applications. These include their use in detectors for electromagnetic radiation over a wide range of frequencies, including optical frequencies$^1$-$^4$. Much of the reported work attributes the observed photoresponse to bolometric effects in which the film is heated by the incident radiation. Some authors have attributed some of their observations, particularly for grainy films with wide transitions, to non-bolometric phenomena but these interpretations have not been universally accepted.

We report here our observations on the photoreponse of two different YBa$_2$Cu$_3$O$_{7-\delta}$ films to visible radiation. One film was epitaxial and had a smooth morphology while the other had a mixed orientation to the substrate and was quite grainy. Both of these films had comparable critical temperatures and transition widths after patterning into test structures.

2. EXPERIMENTAL PROCEDURES

The sequentially evaporated films were deposited by electron beam evaporation. Details on the formation of these films have been reported previously$^1$-$^3$ but the main parts of the process will be reviewed here. The films were formed from Cu, Y and BaF$_2$ deposited in that order. Five layers of each were deposited for a total of
fifteen layers. SrTiO$_3$ substrates were used. Following deposition the films were annealed in a hot wall tube furnace to form the superconductor. The samples were slowly pushed into the preheated furnace over a five minute period. They were annealed at 900°C for 15 min. The temperature was lowered to 450°C at -2°C/min and held there for 6 hr. Finally the temperature was lowered to room temperature at approximately 2°C/min. The ambient was oxygen bubbled through room temperature water during the high temperature anneal and dry oxygen at all other times. The thickness of the sequentially evaporated film for these experiments was 0.5 μm. Films produced by this procedure typically have a critical temperature of 85 K, a granular morphology with a "basket weave" texture and mixed orientation.

The epitaxial film was formed by laser ablation from a YBa$_2$Cu$_3$O$_{7-δ}$ target. During deposition the substrate was heated to 630°C and the chamber pressure was 170 mtorr oxygen. The wavelength of the laser was 248 nm, the energy density was 1.5 J/cm$^2$/pulse and the pulse rate was 4 per second. The laser beam was incident on the target at 15° from the normal. After deposition the oxygen pressure was raised to 1 atm and the temperature was lowered to 450°C at -2°C/min. It was held there for 2 hr and then slowly lowered to 250°C. The film had a thickness of approximately 0.2 μm and a smooth morphology.

For the photoresponse measurements the films were patterned into an "H" shaped detector. The photolithographic procedure employed KTI 752 negative photoresist. The films were etched in 1:100 bromine:ethanol (molar). The bridge region of the "H" was 50 μm wide.

Electrical contacts were made to each of the four legs of the "H." The metalization for the contacts consisted of 0.7 μm of Ag and 0.3 μm of Au. The Au top layer was used to facilitate wire bonding. The contacts were patterned through a chlorobenzene assisted lift-off procedure employing positive photoresist. Following deposition, the contacts were annealed at 500°C in oxygen.

Figure 1. Scanning electron micrograph of a detector fabricated from a sequentially evaporated superconducting film.
Figure 1 shows a scanning electron micrograph of a detector made from the sequentially evaporated film. The darkest region is the superconductor. The four contacts are also visible on the legs of the "H". Also evident in this micrograph are many small blisters in the film. These blisters occasionally form on the sequentially evaporated films during the annealing procedure. They do not occur on all samples formed with nominally the same procedures but unfortunately did form on this sample. Electrical measurements showed continuity and measurements on this sample were carried out in spite of these defects. Figure 2 is a higher magnification micrograph of the bridge region of the same detector. The granular basket weave morphology is apparent. Notice that the basket weave structure is not apparent on the blister, where the film has come out of contact with the substrate.

Figure 2. Higher magnification scanning electron micrograph of the bridge region of the sequentially evaporated detector.

The bridge region of a detector formed from the epitaxial laser ablated film is shown in Figure 3. The film is very smooth, although there are some small particles on the surface. (The large particles are dirt or dust.)

For measurement of the resistance-temperature characteristics and photoresponse of the samples, the substrates were mounted onto the sample holder of a closed cycle He cryostat. Gold wire bonds provided the electrical connections between the four contacts on the detectors and pins on the sample holder. Two of the contacts, one on each side of the bridge were used for current biasing while the other two were connected to either a voltmeter, for resistance measurements, or a lock-in amplifier, for photoresponse measurements.

For the photoresponse measurements the sample was illuminated with chopped He-Ne laser radiation. The light was focused onto the bridge region of the detectors through a window in the housing of the cryostat. To position the beam on the bridge for initial measurements, the temperature was adjusted to approximately the midpoint of the resistive transition. The detector was then illuminated and the position and focus of the beam was adjusted to maximize the detected signal. Additional comments on this will be made later in the paper. The diameter of the focused beam was small.
After positioning the beam, the photoresponse was measured as the temperature of the sample was varied. The measurements reported in this paper were made with the illumination chopped at 400 Hz and the sample biased at 100 μA. A few measurements of the photoresponse as a function of chopping frequency up to 4 KHz were made with the temperature fixed. The signal was found to decrease by approximately 40% as the frequency increased over this range. Measurements were also made at several lower bias currents. The response was found to scale with current and those results will not be further reported here.

3. RESULTS

The resistance (R) in ohms, dR/dT in ohms/K and measured signal in μV for the detector made from the epitaxial film are shown as a function of temperature from 70 to 90 K in Figure 4. The curve for dR/dT has been multiplied by a factor of five so that it could be plotted on the same scale as the others. The temperature of the sample was held at 79.4 K during optimization of the beam position. During the measurement it was varied down to approximately 12 K. The only observed response was in the range plotted in the figure. (Neglecting the small and nearly constant response at higher temperatures.) The peak of the photoresponse coincides well with the peak in dR/dT and the two curves agree well. The slight displacement of the two peaks is within the uncertainty in thermometry as the resistance-temperature characteristic and the photoresponse were not measured simultaneously. The agreement indicates a bolometric photoresponse.

Figure 5 is a graph of the resistance, dR/dT and the initial measurement of the
Figure 4. Resistance (squares), $5 \times \frac{dR}{dT}$ (triangles) and photoresponse (circles) of the laser ablated superconducting film. 400 Hz chopping frequency, He-Ne laser illumination, 100 $\mu$A bias.

photoresponse signal for a detector made from the sequentially evaporated film. The curve for $\frac{dR}{dT}$ has been multiplied by a factor of 10. The temperature of this sample was held at 78.5 K during optimization of the beam position. As with the epitaxial sample, the temperature was varied down to approximately 12 K and no photoresponse other than that shown in this figure was observed. The agreement between the photoresponse and $\frac{dR}{dT}$ is poor. The maximum of the signal is displaced from the maximum of $\frac{dR}{dT}$ by approximately 3 K to a higher temperature. Notice however that there is a shoulder on the peak of the signal at approximately the temperature of the peak in $\frac{dR}{dT}$ and that there appears to be a shoulder on the peak of $\frac{dR}{dT}$ at the peak in the signal.

In speculating on the possibility of experimental problems that might explain this result, a rough calculation showed that thermal expansion of parts in the cryostat could shift the sample on the order of 10 $\mu$m relative to the focused laser beam. The sample was remeasured and, to correct for motion due to expansion, the beam was repositioned every one to two degrees of temperature change.

The remeasured response is plotted in Figure 6. The resistance-temperature characteristic was also remeasured using finer temperature increments. Note that while $R$ and $\frac{dR}{dT}$ are plotted on the same scale in this figure as in Figure 5, the signal is divided by a factor of 2. The measured signal voltage was nearly a factor
Figure 5. Resistance (squares), $10 \times \frac{dR}{dT}$ (triangles) and initial measurement of the photoresponse (circles) of the sequentially evaporated superconducting film. 400 Hz chopping frequency, He-Ne laser illumination, 100 $\mu$A bias.

of three larger when the position of the beam was optimized as the measurement progressed. The agreement between the peak positions was still approximately the same. In addition a shoulder still appeared to exist on the signal peak at about the temperature of the peak in $\frac{dR}{dT}$ and on the $\frac{dR}{dT}$ curve at about the peak in the signal.

4. DISCUSSION

The observed photoresponse of the granular sequentially evaporated film can be explained with two assumptions: 1) That the film is spatially nonuniform with different critical temperatures in different regions, and 2) That the laser beam was not uniformly illuminating the entire bridge area. With these assumptions, as the signal is optimized at a given temperature, the laser beam can be positioned at a location on the film that has a locally high $\frac{dR}{dT}$, even if it doesn’t make a dominant contribution to the total resistance of the film. This can be particularly true if the size of the laser spot is comparable in size to the non-uniformities.

Several simple one dimensional simulations were made to explore this possibility. Three of these will be presented here. In the first two a one dimensional detector was assumed to consist of a series combination of two regions, one with a
transition centered about 83 K and another with a transition centered about 79 K. The higher temperature transition was assumed to be broader. The illumination was assumed to result in a temperature increase that had a Gaussian distribution along the detector. The resistance, dR/dT and the signal for two beam positions are plotted in Figures 7 and 8. In Figure 7 the position of the beam was optimized at 77 K resulting in a strong response due to the low Tc region of the detector while for Figure 8 it was optimized at 87 K resulting in a strong response due to the high Tc portion of the detector.

A slightly more sophisticated simulation was also made. The one dimensional detector was assumed to consist of a narrow region of high Tc with broader regions of lower Tc on either side. The simulation was then run with the beam position re-optimized at 1 K intervals. The result in Figure 9 shows many of the features observed in the measurement on the sequentially evaporated film. The peak in the photoresponse is at a higher temperature than the peak in dR/dT and each peak has a shoulder that corresponds with the other, although the shoulders are much stronger here. The cusp in the signal between the peaks results from the sharp boundary between the regions. The parameters of the model could be adjusted to give a better reproduction of the measured data but this simulation demonstrates that a bolometric response in a nonuniform film can explain the observed signal.
Figure 7. Simulated resistance, $5 \times \frac{dR}{dT}$ and signal of a nonuniform detector. Beam position optimized at 77 K.

Figure 8. Simulated resistance, $5 \times \frac{dR}{dT}$ and signal of a nonuniform detector. Beam position optimized at 87 K.
Figure 9. Simulated resistance, 5 x dR/dT and 5 x signal of a nonuniform detector. Beam position re-optimized at 1 K intervals.

5. CONCLUSIONS

The photoresponses of a laser ablated epitaxial film and a granular sequentially evaporated film of YBa$_2$Cu$_3$O$_{7-\delta}$ have been measured. For both films the only observed signal occurred for temperatures near the transition temperature. For the epitaxial film there was good correspondence between the measured signal and the temperature derivative of the resistance indicating that the photoresponse was bolometric in nature. The photoresponse of the granular films did not coincide as well with dR/dT, however simulations based on the assumption that the film is nonuniform lead to the conclusion that nonuniformities, coupled with a bolometric effect, are sufficient to explain the observations.

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REFERENCES


In this paper we report on the response of thin films of YBa$_2$Cu$_3$O$_{7-x}$ with either a very grainy or a smooth epitaxial morphology to visible radiation. SrTiO$_3$ substrates were employed for both types of films. The grainy films were formed by sequential multi-layer electron beam evaporation while the epitaxial films were formed by laser ablation. Both films were patterned into +H shapes detectors via a negative photolithographic process employing a Br/ethanol etchant. The bridge region of the +H was 50 $\mu$m wide. The patterned films formed by laser ablation and sequential evaporation had critical temperatures of 74 K and 72 K respectively. The bridge was current biased and illuminated with chopped He-Ne laser radiation and the voltage developed in response to the illumination was measured. A signal was detected only above the critical temperature and the peak of the response coincided with the resistive transition for both types of films although the correspondence was less exact for the grainy film. The details of the responses and their analysis are presented.