OPTICAL AND MICROWAVE DETECTION USING Bi-Sr-Ca-Cu-O THIN FILMS

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Abstract

Recent progress at the Johns Hopkins University Applied Physics Laboratory (JHU/APL) in the development of optical and microwave detectors using high temperature superconducting thin films will be described. Several objectives of this work have been accomplished, including: deposition of Bi-Sr-Ca-Cu-O thin films by laser ablation processing; development of thin film patterning techniques, including in-situ masking, wet chemical etching and laser patterning; measurements of bolometric and non-bolometric signatures in patterned Bi-Sr-Ca-Cu-O films using optical and microwave sources, respectively; analysis and design of an optimized bolometer through computer simulation; and investigation of its use in a Fourier transform spectrometer. This paper will focus primarily on our fabrication developments and results from the measurement of the bolometric and non-bolometric response.

Introduction

The development of high temperature superconducting (HTSC) thin film devices for electro-optical and radio-frequency sensors is probably one of the most likely near-term outcomes of recent breakthroughs in the field of superconductivity. Among the many potential devices we have considered at APL passive thin-film devices such as detectors of electromagnetic radiation are favored. We are investigating near-term application of HTSC detectors to laboratory instrumentation for microwave and infrared spectrometry. Response times are on the order of milliseconds to microseconds, depending on the detection mode and wavelength. Operating temperatures are 77 K for bolometric detection, however, the relative sensitivity of the non-bolometric mode of detection requires operation below 77 K.

HTSC materials are expected to exhibit excellent performance as quantum (Josephson junction) radiation detectors, in which case they should operate at extremely high frequencies reaching into the far-infrared band, limited ultimately by the very high superconducting energy gap found in these new materials (≤ 50 meV). Ideal sensitivity should also be good for these type of detectors (≤ 10^{15} photons cm^{-2} sec^{-1} in a 1 kHz bandwidth for a 10^{-4} cm^{2} detector area, as calculated by Forrester and Talvacchio), but it will be worse than low temperature superconducting (LTSC) devices (by the square root of the temperature ratio for noncoherent detection). Fabrication of prescribed (ideal) Josephson junctions is, however, very difficult because the junction size must be on the order of the coherence length, which is extremely short (≤ 20 nm) in these materials. Granular film (multiple Josephson junction) detectors, on the other hand, may, be competitive as detectors, as suggested by Wolf.

HTSC bolometers have been proposed because they are easy to fabricate. They operate on the principle that incident radiation of virtually any wavelength will induce a resistive transition from a virtual superconducting state to normal state in a thin piece of superconducting film. The substrate must be coupled to a thermal reservoir and have a low heat capacity to yield the best possible response to...
incident radiation in the shortest possible response time. Thus there is a tradeoff between response time and sensitivity in bolometric (type II) detectors, which limits their performance relative to ideal quantum detectors. By making the HTSC bolometric element very small and impedance matching it to an efficient antenna structure, better performance should be obtained. Calculated ideal sensitivity (measured by noise equivalent power (NEP)) is in the range of 1-20 x 10^{-12} W Hz^{-1/2}.

We report here results of our investigations of the bolometric and non-bolometric modes of detection in Bi-Sr-Ca-Cu-O thin films. We find that both detection modes appear in the same sample under mutually exclusive conditions (bolometric with light and nonbolometric with microwaves). Konopka, Jung, et al 5-7 have measured the microwave response of Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O films and clearly observed similar behavior in Y-Ba-Cu-O films. Our measurements indicate that the non-bolometric mode of detection obviates the thermal response-time tradeoff of the bolometric mode, a result which offers promise for application to laboratory instrumentation. Recent progress will be described, including: deposition of Bi-Sr-Ca-Cu-O thin films by laser ablation processing 8,9; development of several thin film patterning techniques, including in-situ masking, wet chemical etching, and laser patterning; and measurements of bolometric and non-bolometric signatures in patterned Bi-Sr-Ca-Cu-O thin films using optical and microwave sources. Possible mechanisms for the non-bolometric detection mode are reviewed and future measurements to determine performance limits are discussed.

**Deposition and Patterning Techniques**

Among the several laser beam processing techniques developed at APL, the deposition of thin films by laser ablation processing (LAP) was the first and most important. In that method (Fig. 1) a pulsed beam from an excimer laser is focused onto a pressed pellet of superconducting oxide that is mounted in a vacuum cell, and ablated material is collected on a substrate a few centimeters away tilted at ~ 30° relative to the target. The excimer laser operates at 193 nm, in the ultraviolet region of the spectrum, with a pulse frequency of nominally 10 Hz and an energy of approximately 150 mJ per pulse. During the deposition, the target is translated stepwise so that the beam focal spot of approximately 0.5 mm$^2$ resides at any single location on the target material for 30-100 seconds, which improves the stoichiometry of the film. In addition, the substrate can be heated, and new samples or new targets introduced without breaking vacuum by using a carousel sample support. Substrates can also be translated during deposition to improve thickness uniformity.

![Diagram](attachment://diagram.png)

**Fig. 1:** Laser ablation processing cell used to deposit superconducting thin films.
The laser ablation processing technique has been used to deposit thin films of La-Sr-Cu-O, Y-Ba-Cu-O, and Bi-Sr-Ca-Cu-O on a variety of substrates that include fused quartz, zirconia buffered quartz, crystalline silicon, sapphire and oriented crystals of strontium titanate, zirconium oxide (ZrO₂), and magnesium oxide (MgO) 9-10. The quality of the film depends on the substrate as well as deposition parameters, such as substrate temperature during deposition, and the post-deposition annealing temperature.

For the optical and microwave results reported here the Bi-Sr-Ca-Cu-O films were deposited on MgO and ZrO₂. An example of one of our early Bi-Sr-Ca-Cu-O films on ZrO₂ is shown in Fig. 2, showing the texture and granularity of these films. Film thickness was 1-2 μm and average grain size was estimated at 3-5 μm. A "slow" annealing schedule was established to achieve better film properties, as measured by resistivity. The annealing schedule consisted of a 25°C to 810°C ramp-up for 1 hour, a soak at 810°C for 10 minutes, followed by a 810°C to 25°C ramp-down for 3 hours. Earlier measurements of the resistive transition curve and the magnetically modulated microwave absorption (MAMMA) response reveal a transition temperature (T_c) of ~ 77 K. From this earlier work 9,10 the benefit of heating the substrate during deposition was clearly established. Heating the substrate makes the resistive transition narrower and helps to achieve smoother and more oriented films.

Fig. 2: Optical microscope photograph of a Bi-Sr-Ca-Cu-O thin film under 2500X magnification showing grain sizes on the order of 3-5 μm. This sample was used for subsequent testing.

Several patterning techniques have been tried, including in-situ deposition masking, wet chemical etching, and laser patterning. These tools are available individually or in combination to pattern complex planar structures, such as meanderlines and microstrip. In-situ masking was our earliest attempt at patterning, and it was used to achieve 625 μm linewidths, which are sufficient to make simple detector elements and striplines. A 50 μm thick Kovar mask was used. The limitations of using a shadow mask are that line-widths are limited to 50 μm or more, and complex patterns cannot be made. Thus procedures for wet-chemical etching of Bi-Sr-Ca-Cu-O films were developed. These procedures incorporated a Shipley 21822 photoresist and a selection of acid etches. Choice of the acid etch depends on film type (Y-Ba-Cu-O or Bi-Sr-Ca-Cu-O) and whether the film had been previously annealed. Then linewidths of 1 μm can be achieved. An example of wet chemical etching is shown in Fig. 3, illustrating a prototype meanderline element to be used as a bolometer.
To achieve finely controlled, complex patterns on very small substrates (~1 mm x 1 mm) a laser patterning system was developed as shown in Fig. 4. Similar efforts by other groups have been applied to Y-Ba-Cu-O films. Rothschild, et al. and Liberts, et al. show that by using an oxygen atmosphere while exposed to a laser beam the film can be locally heated and consequently annealed. By using a reducing atmosphere (including nitrogen) an initially superconducting film can be driven back into the normal phase. For our initial testing we increased the laser power to a relatively low threshold level so that the film could actually be etched in ambient air. Thus finely sketched (computer controlled) patterns can be produced on substrates too small to be patterned by conventional wet chemical etch procedures. This approach employs an Argon ion laser operated at 514.5 nm with a ≤ 10 μm spot size and 7 x 10^5 W/cm² maximum irradiance on the substrate. Laser modulation is enabled electro-mechanically (or acousto-optically). Substrate positioning is achieved with a computer-controlled x-y translation stage, which has a 0.1 μm stepping resolution. A vacuum cell with multi-axis optical access is used to house the film. Partial pressure of oxygen and/or nitrogen from ~ 100 mTorr to one atmosphere can also be maintained during operation, in order to explore local annealing of Bi-Sr-Ca-Cu-O films. The sample is monitored under reflected ambient light using a charge coupled device (CCD) camera and telephoto/zoom lens combination, and the transmitted power is measured with a photodetector.
In a new on-going effort, a meanderline HTSC bolometer was developed using the LAP cell and wet chemical etching as shown in Fig. 3. A computer program for calculating bolometer response was developed to support the design of the bolometer, which included a noise budget. The particular design pictured in Fig. 3 (5 x 5 mm) is not the smallest that can be achieved and will be reduced by a factor of 5 to achieve our desired performance goal. The requirement to reduce substrate size in order to reduce heat capacity will dictate the use of our laser patterning system. Our goal is that the HTSC detector should be at least as sensitive as the pyroelectric detector (deuterated triglycerine sulfide (DTGS)) and cover a broader spectral range, extending beyond the DTGS range of 0.25-500 μm. This design will be isolated thermally to achieve a desired response time (τ = 10 msec) and interfaced to a low temperature JFET preamp to achieve low noise (NEP = 10^-10 W Hz^-1/2).

To demonstrate the use of the laser patterning system, a simple meanderline pattern was sketched on a Y-Ba-Cu-O thin film, as shown in Fig. 5. The laser was guided by computer between the two closest contacts (spaced apart by ~ 1600 μm). The linewidth of the meanderline was 400 μm, and the furrows created by the laser beam were ~ 10 μm wide. This sample and another were tested and results indicate that they have good electrical isolation, increased resistance over the unpatterned sample (by one to two orders of magnitude), and a bolometric response.

![Fig. 5: Simple meanderline element sketched on Y-Ba-Cu-O on MgO using Argon ion laser beam.](image)

**Optical and Microwave Detection Measurement Results**

Many investigators are interested in developing a practical high temperature superconducting detector. APL is interested in detectors that can be used in lieu of a conventional bolometer in a laboratory Fourier transform spectrometer (FTS). Testing and design work are underway to ultimately integrate a very small meanderline element, similar to the prototype shown in Fig. 3 with a low-temperature JFET amplifier (and other electronics), which will yield signal levels compatible with the A/D converters on our FTS. We are also interested in developing detectors for similar microwave instrumentation. In a preliminary effort described below we have measured the optical and microwave response of simpler patterned Bi-Sr-Ca-Cu-O films using a standard four-point probe configuration.

Two of the samples tested in these preliminary measurements consisted of a small section of a patterned Bi-Sr-Ca-Cu-O film deposited in the LAP cell on a single crystal MgO substrate at 300 °C and on single crystal ZrO₂ at room temperature. Film thicknesses were 2 μm and 1.3 μm, and film areas were 2000 by 600 μm and 1500 by 370 μm, respectively. These samples were shadow-masked during deposition and annealed as previously described. Silver epoxy contacts were placed on the sample and annealed-in during film annealing, a procedure that almost always eliminates contact resistance problems. Typical
contact resistances varied from a few ohms at room temperature to approximately 0.1 ohm at 13 K. In addition, we find that the samples can be thermally recycled many times with little degradation of the contacts or the intrinsic film properties, and they can be re-used after a long (~ 6 month) shelf-life.

For optical detection, a 4 mW, 633 nm helium-neon laser was chopped at 26 Hz and focused onto the center of the sample. For microwave detection, a 9 GHz microwave signal was generated with a microwave oscillator and square wave modulated at 40 Hz with a PIN diode modulator. The signal was then amplified and fed into an X-band horn positioned directly in front of the sample. In both cases the induced output voltage from the sample was synchronously detected with a lockin amplifier. See Fig. 6(a) and 6(b) for details.

Fig. 6: (a) Bolometric response measurement set-up and (b) microwave response measurement set-up, shown adjacent to cold head and post detection electronics.

Results for the optical detection experiment shown in Fig. 7 indicate a response peak located at the center of the transition region. From standard bolometric theory, it is known that the bolometric response is proportional to the derivative of the resistance curve. Calculated derivatives of the resistance curves correlate well with the measurements, as shown. In addition, measurements of the lockin response versus chopper frequency shown in Fig. 8 indicate response times (~ 5.7 and 11.5 msec) consistent with a thermal response mechanism dominated by substrate characteristics.

Fig. 7: Optical response, resistance, and dR/dT versus temperature of Bi-Sr-Ca-Cu-O sample. Bias current was set to a relatively high value (1.04 mA).
Unlike the optical response, the peak in the microwave response is located (in temperature) in the region of the resistive tail well-below $T_c$ as shown in Fig. 9 and clearly separated from the optical bolometric response peak shown previously in Fig. 7. This implies that the microwave response is non-bolometric. As expected, the width of the resistive tail increases with increasing microwave power, and the peak of the response increases until saturation, as shown. In addition, lockin response to microwaves does not rolloff with chopper frequency (up to 50 kHz) as did the optical response, implying the microwave response mechanism is not thermal.

![Graph showing microwave response versus temperature for different microwave powers](image)

**Fig. 8:** Optical bolometric response versus chopper frequency for two Bi-Sr-Ca-Cu-O samples.

![Graph showing microwave response versus temperature](image)

**Fig. 9:** Microwave response of Bi-Sr-Ca-Cu-O sample versus temperature for different microwave powers. The inset is a plot of the peak response vs microwave power.

Noise voltage measurements were also taken with the lockin amplifier using a low noise pre-amplifier and biasing with a battery source. Similar microwave noise emission results were observed by Konopka, et al in Y-Ba-Cu-O films. For our measurements, the equivalent noise bandwidth was set to 1 Hz and the chopping frequency was set to 40 Hz. Even with no illumination the sample has a response, and it is in the resistive tail region as shown in Fig. 10. In addition, peak excursions of the noise voltage observed in the region of the peak were much higher than the RMS noise voltage levels. This behavior would be expected if individual transient fluctuations occur in the film over very short time intervals, perhaps associated with flux motion and dissipation induced by bias current flow.
Fig. 10: Noise voltage versus temperature for Bi-Sr-Ca-Cu-O sample for various bias currents. The R(T) curve is shown for comparison.

Models of Response Mechanism

Several hypotheses have been put forward to explain the microwave response peak. It could be ascribed to flux-flow within a weakly-coupled granular weak-link array. Some of the evidence used to support this vortex-antivortex dissociation mechanism (also known as the Kosterlitz-Thouless model) includes the presence of a resistive tail below the superconducting transition\textsuperscript{20}, which we observe. In addition to evidence for this type of response in granular Y-Ba-Cu-O films noted by Culbertson, et al\textsuperscript{18}, Zeldov et al\textsuperscript{21} have observed a non-bolometric response in epitaxial Y-Ba-Cu-O films, and they explain their results on the basis of photoenhanced flux creep. This mechanism, however, is not likely to be the cause of the optical response seen in our granular films since the peak location and response time are consistent with the thermal (bolometric) response, but it may be a mechanism for the microwave response. In fact, earlier work by Voss et al\textsuperscript{22} in granular aluminum films demonstrated an almost identical behavior to our microwave results. Recent results of Konopka, et al\textsuperscript{5-6} and Jung, et al\textsuperscript{7} also reveal similar behavior in Y-Ba-Cu-O films, as well as Bi-Sr-Ca-Cu-O films, but the results for the noise emission in Bi-Sr-Ca-Cu-O were barely detectable. We observe clear indications of microwave noise emission in our samples.

The responsivity of the bolometric detection mode using HeNe laser light (633 nm) was calculated from the experimental parameters. Values of 5 mV/W (Bi-Sr-Ca-Cu-O on MgO) and 250 mV/W (Bi-Sr-Ca-Cu-O on ZrO\textsubscript{2}) were obtained for a chopper frequency of 20 Hz. In contrast, the responsivity of the nonbolometric mode using a X-band (9 GHz) horn was calculated to be 20-120 V/W (Bi-Sr-Ca-Cu-O on MgO) independent of chopper frequency up to the limit set by our current experimental apparatus (50 kHz), which is considerably higher than the bolometric mode. Of course, the bolometric mode was not optimized for maximum responsivity in these preliminary samples. Nor was the microwave response necessarily optimized, but the means to optimize it are not as clear because the exact mechanism is not known. Gallop, et al\textsuperscript{23} offer a plausible explanation based on their measurements of the differential resistance (dV/dI) of Y-Ba-Cu-O samples and the idea that microwave radiation synchronizes with the weak fluxon lattice likely present in the granular film. Fluxons move, creating a Faraday voltage due to the Lorentz force and a consequent dissipation measured by dV/dI. If the rate of passage of fluxons across the sample is proportional to the microwave frequency (f) then the corresponding voltage drop across the sample is $V = n\Phi_0 f$, where $n$ is an integer and $\Phi_0$ is the flux quantum. This result is similar to the Josephson relation for Shapiro steps, except that the resulting dV/dI indicates a dissipation rather than a supercurrent at voltage values proportional to frequency. We strongly expect to see similar results in our samples as we measure the frequency dependence of the nonbolometric response.
Summary

Deposition and patterning techniques have been developed based on the use of lasers, which are very effective and flexible in developing practical HTSC thin film detectors. We are continuing to refine these techniques to implement detectors for laboratory-based infrared and microwave spectroscopy. Measurements of Bi-Sr-Ca-Cu-O patterned thin films reveal the presence of optical bolometric and microwave nonbolometric responses in the same samples. The nonbolometric response appears to be a faster, more sensitive mechanism than the bolometric response by over three orders of magnitude in these preliminary results, but further design refinements and measurements will determine the actual relative performance. Measurements will be made by varying the microwave frequency and static applied magnetic field. These proposed measurements should help resolve issues regarding the response mechanism. We also plan to use fast rise-time microwave excitation to attempt to determine the ultimate bandwidth.

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References


