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Cryogenic performance of semiconducting Y–Ba–Cu–O for infrared detection

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Abstract. At room temperature, YBCO possesses a relatively high temperature coefficient of resistance, -3.5% K^{-1}, and a high pyroelectric coefficient, up to 20 μC cm^{-2} K^{-1}. In this work, we report on the cryogenic characterization of semiconducting Y–Ba–Cu–O thin films on substrates in response to 100 fs optical pulses at 810 nm and suspended thin film pixels in response to chopped broadband infrared radiation. The temperature dependence of the zero-bias pyroelectric response of the detectors is reported.

1. Introduction

Semiconducting Y–Ba–Cu–O (YBCO) has been demonstrated to be attractive as a thermal infrared detector operating as both a bolometer [1] and a pyroelectric detector [2, 3]. In this paper, the pyroelectric behaviour of YBCO is investigated at cryogenic temperatures in response to femtosecond optical pulses and chopped broadband infrared radiation.

The thermoelectric behaviour of crystalline metallic ($x \sim 1$) and semiconducting (oxygen deficient, $x \leqslant 0.5$) $YBa_2Cu_3O_{6+x}$ (YBCO) and similar compounds in the normal state [4–15] has been studied extensively in recent years. Very recently, thermoelectric behaviour has been investigated in amorphous, semiconducting YBCO thin films [16]. The latter studies were motivated by the application of YBCO as an uncooled thermal detector for infrared and other radiation or solar cells. The thermoelectric behaviour has been attributed to the pyroelectric effect [5, 16, 17], Seebeck effect [6] and photovoltaic effect [18] in Y–Ba–Cu–O. In addition, piezoelectricity has been reported in Y–Ba–Cu–O. Although pyroelectricity and piezoelectricity should be forbidden owing to the centrosymmetric crystal structure of Y–Ba–Cu–O, Mihailovic and Foster [17] postulated the symmetry to be broken by the coupling of the hole to the apex oxygen of a CuO_2 plane.

2. Background

The pyroelectric effect refers to the temperature dependence of the spontaneous electric polarization resulting in the variation of the surface charge and the associated flow of a pyroelectric current [19]. A voltage results from the pyroelectric current flowing through the equivalent impedance of the detector and amplifier recording the signal.

In the case of a chopped or a sinusoidal optical intensity, the voltage responsivity R_v is given by [19]

$$R_v = \frac{\eta}{C_{th}} \frac{pA}{C} \frac{\omega\tau_e}{(1+\omega^2\tau_e^2)^{1/2}} \frac{\tau_{th}}{(1+\omega^2\tau_{th}^2)^{1/2}} \qquad (1)$$

where p is the pyroelectric coefficient of the detector material, A is the area of the detector sampling the change in the surface charge, η is the absorptivity, ω is the modulation frequency of the light, τ_e ($= RC$), is the electrical time constant determined by the equivalent circuit of the detector and preamplifier and τ_{th} is the thermal time constant of the detector ($\tau_{th} = C_{th}/G_{th}$ with thermal mass C_{th} and thermal conductance G_{th} to the substrate). Under pulsed laser illumination with the optical pulse duration much less than both the electrical and the thermal time constants; the optical pulse can be approximated as an impulse, yielding the time-varying voltage [15]:

$$v_A(t) = \frac{K\tau_e}{\tau_{th}-\tau_e}\left[\exp\left(-\frac{t}{\tau_e}\right) - \exp\left(-\frac{t}{\tau_{th}}\right)\right] \qquad (2)$$

where $K = pA\eta\Phi/C_{th}C$ with Φ as the incident optical power.

3. Experimental details and results

The Y–Ba–Cu–O thin films used in these experiments were fabricated by rf magnetron sputtering at ambient temperature. Films deposited in this manner are polycrystalline to amorphous with a typical stoichiometry of 1:0.5:2.2:4.6, as determined by a wavelength-dispersive electron probe. As deposited, the films exhibit semiconductor-like resistance versus temperature characteristics with a temperature coefficient of resistance of -3 to -4% K^{-1} and resistivity of 5–10 Ω cm.

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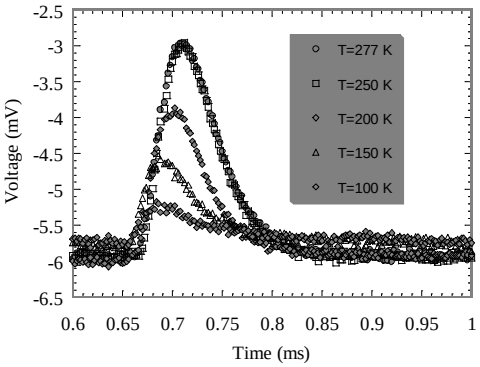

Figure 1. Unbiased voltage response of YBCO thin film illuminated by 100 fs, 810 nm laser pulses at different temperatures.

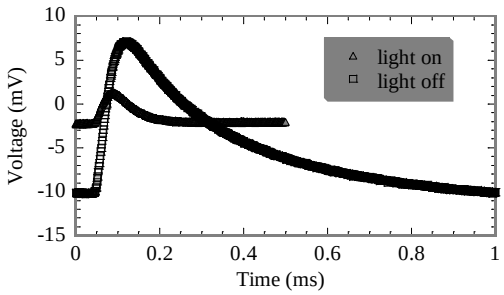

Figure 2. Voltage response of YBCO at 300 K when illuminated with and without illumination by CW light in addition to 100 fs laser pulses. The average power of the laser pulses was 200 μW.

The pulsed laser experiments employed 200 nm thick films deposited on oxidized silicon wafers. Silver (Ag) electrodes were fabricated onto the YBCO film in a microstrip transmission line geometry (100 μm wide) to allow for the propagation of high-speed electrical signals. A 20 μm wide gap was patterned in the Ag microstrip to allow for optical illumination of the YBCO and form a 'photoconductive switch' geometry. However, no bias was used during the measurements. The sample was mounted on a substrate holder providing the conducting ground plane for the microstrip. Aluminium (Al) wires were ultrasonically bonded to the silver microstrips to provide electrical contact. The substrate holder was mounted on a cold finger in an evacuated cryostat. The voltage response was measured directly with a 400 MHz bandwidth oscilloscope. The sample was illuminated by 100 fs optical pulses at 810 nm with a pulse repetition rate of 1 kHz. The optical intensity was varied with neutral density filters. The optical spot size is estimated to be 20 μm at the sample.

Typical unbiased voltage responses from the pulsed laser illumination are shown in figure 1. The voltage signal corresponds to the change in voltage across the gap in the microstrip. In this geometry, the Ag microstrips sample the change in the surface charge due to the temperature dependence of the spontaneous polarization. The nature of the voltage response was relatively independent of the location of the excitation. The polarity and general shape of the voltage signal did not depend on whether the gap was illuminated directly, the silver electrodes were illuminated or the beam was positioned about 100 μm beside the gap and silver microstrips. However, the magnitude of the voltage signal depended upon the position of illumination. We believe that this indicates that the voltage response was thermal in origin, resulting from the heating by the optical pulse. The data were fitted to equation (2), yielding distinct time constants for the fall and rise of the voltage signal. The rise time constant was observed to decrease with decreasing temperature while the fall time constant increased with decreasing temperature. The rise time constant is interpreted to be the electrical time constant τ_e of the system while the fall time constant is interpreted as the thermal time constant τ_{th}. The temperature dependence of τ_e is observed to obey an Arrhenius relationship $\tau = \tau_0 \exp(-E_A/k_B T)$ where $E_A = 0.025$ eV is the activation energy, τ_0 is a constant and k_B is Boltzmann's constant. Although τ_e and τ_{th} had similar magnitudes near room temperature, their magnitudes differed by more than an order of magnitude at 100 K. The different temperature dependences indicated that the time constants correspond to intrinsically different processes.

Curiously, in some cases, the fall time constant was observed to change significantly when the sample was also illuminated by a continuous-wave incandescent lamp during the laser pulse excitation. As shown in figure 2, the relaxation, when illuminated by the lamp, was at least one order of magnitude shorter than when the sample was dark. The dependence of the relaxation rate on illumination was not present in most samples. The amplitude of the voltage response also displayed a corresponding dependence on continuous-wave illumination as well as the value of the baseline voltage present between pulse excitations. At this time, the mechanism for the reduction in the relaxation time with continuous illumination is not well understood.

The energy (power) dependence of the voltage response was also investigated. The time constants were found to be independent of the laser pulse energy. The amplitude of the voltage response depended directly on the laser pulse energy, increasing with increased energy. The voltage response was observed to saturate in amplitude at large pulse energies (0.6 μJ). The strength of the laser illumination was sufficient to write a visible pattern in the YBCO film. The amplitude of the voltage response diminished over time from continued laser illumination. Illumination by the laser light altered the YBCO film properties.

The YBCO was fabricated into thermal isolation structures by employing silicon (Si) micromachining techniques. Suspending the detector above the substrate decreases the thermal conductance G and thereby increases the responsivity as predicted by equation (1). 1×10 detector arrays with pixels measuring 40 μm $\times$ 40 μm were produced. A scanning electron micrograph of the detector array is shown in figure 3.

The detectors were fabricated by depositing and patterning Al mirrors on the substrate. The Al mirrors form a resonant cavity with the pixel to enhance optical absorption. A 1.7 μm thick polyimide mesa was patterned as a sacrificial layer upon which the pixels were fabricated above the Al mirrors. The detector pixels were formed by sequentially depositing 400 nm of YBCO and 100 nm of niobium (Nb). The Nb electrodes were patterned by dry

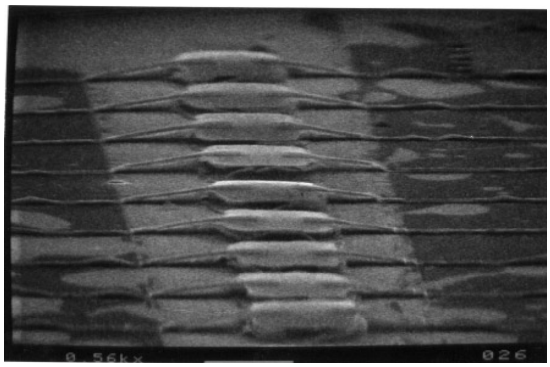

Figure 3. A SEM micrograph showing a 1×10 YBCO detector array suspended above the substrate. The pixel size is $40\ \mu\text{m} \times 40\ \mu\text{m}$.

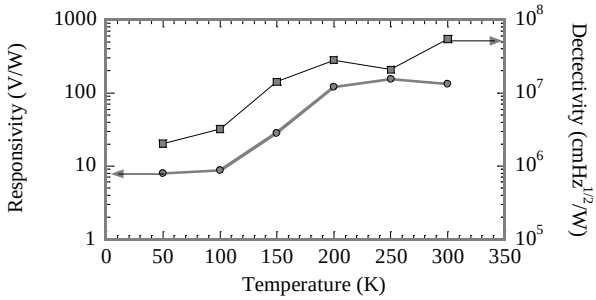

Figure 4. Temperature dependence of unbiased YBCO responsivity and detectivity in response to broadband infrared radiation chopped at 330 Hz.

etching. The YBCO pixels were patterned by etching with Al etch 1:20 concentration. The pixels were over-etched, removing the YBCO from under the Nb electrode arms. The polyimide was then ashed in an oxygen plasma to remove it from under the pixels and suspend the YBCO film above the substrate.

The completed detectors were wire bonded into a package and mounted in a cryostat for characterization. The detectors were illuminated with chopped broadband infrared radiation covering the spectrum from 0.6 to 12 μm. The voltage response was measured using a PAR 113 amplifier and signal analyser to determine the corresponding voltage signal and noise levels. The responsivity was calibrated with an Oriel pyroelectric detector.

The unbiased responsivity and detectivity of the YBCO detector are plotted versus temperature in figure 4. The chopper frequency was 330 Hz, corresponding to a rate where the responsivity was relatively constant with respect to chopper frequency. The detectivity ($D^* = R_V(A\,\Delta f)^{1/2}/V_n$) is determined by the area-normalized signal-to-noise ratio where Δf is the bandwidth and V_n is the noise voltage. The data display relatively constant behaviour with decreasing temperature from room temperature to approximately 200 K. From 200 K to 100 K, the detectivity and responsivity decrease dramatically. The responsivity and detectivity appear to level off below 100 K. The decrease in the YBCO detector responsivity can be attributed to an increase in the thermal conductivity of the supporting Nb electrode arms with decreasing temperature and an apparent decrease in the pyroelectric coefficient with decreasing temperature. The latter can also explain the decrease in the voltage amplitude observed in the pulsed laser experiments. Although the detectivity is near 10^8 cm Hz$^{1/2}$ W^{-1} at room temperature, the detectivity falls to 10^6 cm Hz$^{1/2}$ W^{-1} at 50 K.

4. Discussion and conclusions

This investigation has demonstrated that the semiconducting Y–Ba–Cu–O thin films deposited at ambient temperatures continue to exhibit thermoelectric behaviour at cryogenic temperature. The behaviour is believed to be pyroelectric in origin [15]. The magnitude of the effect decreases with decreasing temperature. Since the YBCO films were strain poled as deposited, the decrease in responsivity with temperature may be attributed to the strain associated with cooling. Other factors such as moving further away from the Curie temperature and changes in thermal conductivity may also be responsible. Poling the samples with the application of an electric field may maintain the magnitude of the pyroelectric behaviour at low temperatures. The detectivity of 10^8 cm Hz$^{1/2}$ W^{-1} measured at room temperature is comparable to other room temperature pyroelectric detectors but less than the 10^{10} cm Hz$^{1/2}$ W^{-1} background noise limit of thermal detectors. The observation of a dependence of the voltage relaxation strongly suggests an electronic role in the thermoelectric behaviour of semiconducting YBCO.

Acknowledgments

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References

[1] Jahanzeb A, Travers C M, Çelik-Butler Z, Butler D P and Tan S G 1997 *IEEE Trans. Electron Devices* **44** 1795
[2] Jahanzeb A, Travers C M, Butler D P, Çelik-Butler Z and Gray J E 1997 *Appl. Phys. Lett.* **70** 3495
[3] Butler D P, Çelik-Butler Z, Jahanzeb A, Gray J E and Travers C M 1998 *J. Appl. Phys.* **84** 1680
[4] Chang C L, Kleinhammers A, Moulton W G and Testardi L R 1990 *Phys. Rev.* B **41** 11 564
[5] Mihailovic D and Heeger A J 1990 *Solid State Commun.* **75** 319
[6] Lengfellner H, Kremb G, Schnellbögl A, Betz J, Renk K F and Prettl W 1992 *Appl. Phys. Lett.* **60** 501
[7] Mihailovic D and Poberaj I 1991 *Physica* C **185–189** 781
[8] Poberaj I and Mihailovic D 1992 *Ferroelectrics* **128** 197
[9] Zeuner S, Prettl W and Lengfellner H 1995 *Appl. Phys. Lett.* **66** 1833
[10] Kwok H S, Zheng J P and Dong S Y 1991 *Phys. Rev.* B **43** 6270
[11] Palti A, Svetchnikov V, Pan V and Snarski A 1995 *J. Appl. Phys.* **77** 2814
[12] Grachev A I, Pleshakov I V, Paugurt A P and Shul'man S G 1996 *Phys. Solid State* **38** 1598

[13] Grachev A I and Pleshakov I V 1997 *Solid State Commun.* **101** 507
[14] Mihailovic D, Poberaj I and Mertelj A 1993 *Phys. Rev.* B **48** 16 634
[15] Butler D P, Celik-Butler Z, Adam R and Sobolewski R 1999 *J. Appl. Phys.* **85** 1075
[16] Gray J E, Celik-Butler Z, Butler D P and Jahanzeb A 1998 *Ferroelectrics* **209** 517
[17] Mihailovic D and Foster C M 1990 *Solid State Commun.* **74** 753
[18] Scott J F 1990 *Appl. Phys. Lett.* **56** 1914
[19] Hanson C M 1997 *Semicond. Semimet.* **47** 123