Infrared response in the 95 to 300 K temperature range of detectors based on oxygen-depleted Y-Ba-Cu-O thin films

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Abstract

In order to develop radiation thermal detectors, oxygen-depleted amorphous YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) thin films were grown at low temperature (< 150°C) on SiO$_2$/Si substrates by direct-current hollow cathode sputtering, which allows to envisage further integration of detectors on a readout electronics silicon chip. Planar and metal/YBCO/metal tri-layer sensing structures were then processed. The near-infrared optical response showed evidence of a transition from bolometric to pyroelectric behavior on planar devices, but it was only pyroelectric for tri-layer devices. They exhibited response level and noise performance competitive with other room temperature infrared bolometers with the possibility to enhance this performance by cooling down to 95 K.

1. Introduction

Apart from the well-known superconducting orthorhombic phase of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) at $\delta \leq 0.4$, oxygen depletion leads to the tetragonal crystal structure of a semiconducting Fermi glass at $\delta \approx 0.5$–0.7 and an insulator at $\delta \approx 1$. An unusual although promising application of YBCO in its semiconducting form can be sought in the field of infrared (IR) uncooled thermal detectors of the bolometer type [1, 2] due to its large temperature coefficient of resistance ($TCR = 1/R \frac{dR}{dT} = -3$ to $-4 \% \cdot K^{-1}$) [3], a figure of merit.

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that compares very favorably with other bolometric sensing materials such as VO\textsubscript{x} (−2 %·K\textsuperscript{−1}) [4] or amorphous SiH (−2.5 %·K\textsuperscript{−1}) [5]. Besides, semiconducting YBCO films can be deposited without substrate heating in amorphous semiconducting form [3], hereafter referenced to as a-YBCO, which makes the integration of this material compatible with already processed signal readout electronics (e.g. a complementary metal-oxide-semiconductor (CMOS) chip). Finally, previous work by Butler et al. [6] has shown encouraging IR detectivity values measured on semiconducting YBCO microbolometers, testifying the low noise potential of this material.

The aim of the present work was to examine two types of a-YBCO bolometric geometries, i.e. planar or tri-layer, and compare their IR detection performance in terms of both responsivity and noise level, with reference to already reported semiconducting devices, mainly designed for room temperature operation. We also wish to show here that the pyroelectric behavior of a-YBCO [7] specifically leads to high performance and fast devices that could pave the way to a new generation of sensing pixels for IR imaging ranging from the near-infrared (NIR) to far infrared (FIR) / THz domains.

2. Device fabrication and experiments

2.1. a-YBCO thin film preparation and structural properties

a-YBCO thin films were prepared by off-axis direct-current (DC) sputtering at low temperature (< 150°C) under a ≈ 67 Pa atmosphere of oxygen and argon in the 45% / 55% flow ratio, and with no further oxygenation step, typically leading to the semiconducting deoxygenated YBCO phase [3]. A 2-inch diameter hollow superconducting target of the stoichiometric YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{6.9} phase (Hitec Materials GmbH) was used. The deposition rate was about 2.5 nm/min. We used p-doped silicon substrates of 380 μm thickness and 15 × 15 mm\textsuperscript{2} area, coated with a thermally grown 500 nm thick SiO\textsubscript{2} layer.

Topographical features of film surface were revealed by atomic force microscopy (AFM) using a Nanoscope® III system (Digital Instruments) in the tapping mode. As displayed in Fig. 1(a), the 360 nm-thick a-YBCO film surface exhibited a granular structure with grains isolated in a seemingly uniform amorphous matrix. The surface roughness was low (≈ 5 nm rms).

2.2. Device fabrication

Two types of device structures were fabricated: a planar structure #SiPL, as sketched in Fig. 1(b), and a tri-layer structure #SiTR, as sketched in Fig. 1(c). For both devices, the technological process started with a bottom contact level, patterned by lift-off using a 10 nm-thick titanium layer and a 170 nm-thick gold layer deposited by e-beam evaporation on the substrate. An a-YBCO film of thickness 250 nm for device #SiPL (360 nm for device #SiTR) was then deposited and patterned using standard optical lithography to define the active area. Finally, for device #SiTR, a top electrode (contact pad + connection line) was defined by lift-off using a 10 nm-thick titanium layer and a 170 nm-thick gold layer deposited by e-beam evaporation.

2.3. Experimental set-up

The optical response was measured at 850 nm wavelength with a VCSEL diode, which output power was modulated electronically. The use of optical density filters allowed incident power on the device to be limited to a few μW. The laser beam was focused onto the device at a diameter of 200 μm. The dynamic response and noise measurements were carried out as a function of the laser modulation.
3. Results and discussion

3.1. DC electrical properties

Both (voltage biased) devices exhibited high values of room temperature resistivity ($\approx 350 \Omega \cdot \text{cm}$). The film resistivity $\rho$ followed an Arrhenius law as a function of temperature $T$: $\rho(T) = \rho_e \exp(E_a/k_B T)$, where $\rho_e$ is a pre-exponential factor, $E_a$ the activation energy and $k_B$ the Boltzmann constant. A value of $E_a = 0.25 \text{ eV}$ ($0.16 \text{ eV}$, respectively) was extracted for device #SiPL (#SiTR, respectively). The corresponding TCR value measured at 300 K for device #SiPL spanned between $3$ and $4 \% \cdot \text{K}^{-1}$ [8], in line with the values reported in literature [1]. The TCR values for device #SiTR spanned from $2 \% \cdot \text{K}^{-1}$ at 300 K to $28 \% \cdot \text{K}^{-1}$ at 80 K, as displayed in Fig. 1(d). The current-voltage characteristic for device #SiTR also exhibited a strong non-linearity due to contacts.

3.2. Planar device optical response

Typical response of device #SiPL at 300 K is shown in Fig. 2(a). It includes both the device signal current $i_S$ and noise current density $i_N$ as well as the voltage responsivity $R_V$ at the transconductance amplifier output, as deduced from the effective power sensed by the device (in the $\mu \text{W}$ range). The response exhibited a low-frequency regular low-pass bolometric behavior ($f_B = 28 \text{ Hz}$ cut-off frequency / $\tau_B = 5.7 \text{ ms}$ time constant followed by $\sim f^{-1}$ decrease) as previously reported [8]. Above $200 \text{ Hz}$, a high-pass behavior was observed ($\sim f^1$ increase followed by $f_p = 55 \text{ kHz}$ cut-off / $\tau_p = 2.9 \mu \text{s}$) that could be assigned to the pyroelectric state of a-YBCO [7]. As shown in Fig. 2(b), the pyroelectric response could be extended to the whole operating frequency range, with $f_p = 80 \text{ kHz}$ / $\tau_p = 2 \mu \text{s}$ by adjusting the beam to an off-centered position and promoting thus a wave guiding effect; this observation could confirm the possible role of the p-doped Si substrate acting as a counter electrode for the inter-contact capacitance.
3.3. Tri-layer device optical response

The voltage responsivity vs. frequency exhibited the purely high-pass trend as observed for the planar structure (pyroelectric behavior) with a cut-off frequency of $f_p = 50 – 60$ kHz ($\sim 3 \, \mu$s time constant). Although a regular bolometric bump could be guessed at room temperature as shown in Fig. 3(a), the response became purely pyroelectric upon cooling. As the tri-layer structure favors capacitive effects, these effects overcome the resistive component that falls exponentially as temperature decreases. Although a slightly improved response is exhibited with the larger DC bias voltage (that can be attributed to a poling effect), the overall response decreases at lower temperatures as displayed in Fig. 3(b), showing that the TCR increase is overcompensated by the decrease in electric coupling efficiency, as reported in [9]. The resulting thermal coefficient of capacitance is $-0.5 \, \%/K$ in the $f^1$ portion. This effect also allows to sample the electric polarization temperature dependence and, after normalizing to the total response, extract the pyroelectric coefficient of a-YBCO as $70 \, \text{nC-cm}^{-2} \cdot \text{K}^{-1}$ at 300 K and $23 \, \text{nC-cm}^{-2} \cdot \text{K}^{-1}$ at 100 K. This order of magnitude is in line with direct thermal balance determinations [6]. The signal-to-noise ratio trend is a decrease at lower frequencies, due to both the pyroelectric response and the $1/f$ noise contribution. It peaks at higher frequencies, showing the interest of a-YBCO for fast response ($\mu$s range) detection.

Fig. 2. (a) voltage responsivity $R_V$, device signal current $i_s$ and noise current density $i_N$ of the planar device biased at $V_{DC} = 8$ V and measured at room temperature; (b) enhancement of the pyroelectric response by shifted illumination conditions (see text).

Fig. 3. (a) optical response (signal current $i_s$) and noise current density $i_N$ of the tri-layer device measured at room temperature and moderate cooling; (b) the same device cooled in the 95 to 200 K temperature range.
3.4. Performance results: synthesis

In the following, we compare our results with respect to both previously reported data on semiconducting YBCO bolometers, and also room temperature data on other IR semiconducting bolometric devices, i.e. amorphous silicon or SiGe compounds and VO$_x$ – based compounds. It should be stressed that straightforward comparison was not always possible because of the different device biasing modes (current or voltage – with or without load resistance) that lead to different readout techniques (voltage or current). Consequently, the performances gathered in Table 1 pertain to results for which the available data would allow adequate conversions. Besides, published results are most often at low modulation frequency, because high performance was sought for imaging at 25 – 30 Hz frame rate. In our case, however, the dominant fast pyroelectric response promotes competitiveness at modulation frequencies above 10 kHz, where high sensitivity values are exhibited (with response times in the microsecond range) by both planar and tri-layer structures. The advantage of the latter resides in its lower access impedance that may allow the input radiation coupling by means of an integrated planar micro-antenna for THz applications [8]. With respect to other semiconductors, the possibility to cool a-YBCO pyroelectric devices with slightly degraded response but with an overall improvement in terms of signal to noise ratio is another advantage.

Table 1. Device performance results in the infrared

| Reference | Material / Device | $^a T$ (K) | $^b R_V$ (V/W) @ f (Hz) | $^c$ NEP (W Hz$^{-1/2}$) | $^d D^*$ (cm Hz$^{-1/2}$ W$^{-1}$) | $^e$ $\tau$ (s) |
|-----------|------------------|-----------|------------------------|------------------|---------------------------|----------------|
| This work | a-YBCO / Planar  | 300       | 100 @ 10               | 1.4 x 10$^8$     | 6.95 x 10$^3$             | 5.7 x 10$^-3$  |
|           |                  |           | 7.1 x 10$^4$ @ 8 x 10$^4$ | 2.8 x 10$^{-11}$ | 3.51 x 10$^8$             | 2 x 10$^{-6}$  |
| This work | a-YBCO / Trilayer| 300       | 2.3 x 10$^3$ @ 10$^5$  | 2.5 x 10$^{-10}$ | 5.88 x 10$^7$             | 2.7 x 10$^{-6}$ |
|           |                  |           | 5.7 x 10$^6$ @ 6 x 10$^4$ | 9.6 x 10$^{-11}$ | 1.54 x 10$^8$             |               |
| This work | a-YBCO / Trilayer| 125       | 8.8 x 10$^3$ @ 10$^3$  | 8.8 x 10$^{-11}$ | 1.69 x 10$^8$             | 3.2 x 10$^{-6}$ |
|           |                  |           | 1.3 x 10$^6$ @ 5 x 10$^4$ | 6.9 x 10$^{-11}$ | 2.14 x 10$^8$             |               |
| [6]       | a-YBCO / Trilayer| 300       | 1.3 x 10$^3$ @ 10$^3$  | n.a.             | 1 x 10$^8$                | 0.1 x 10$^{-3}$ |
| [9]       | a-YBCO / Planar  | 300       | 100 @ 330              | n.a.             | 5 x 10$^7$                | n.a.           |
|           |                  | 150       | 10 @ 330               | n.a.             | 2 x 10$^7$                |               |
| [10]      | a-Si / Planar    | 300       | 1 x 10$^6$ @ 200       | 0.8 x 10$^{-11}$ | 1.5 x 10$^9$              | 2.1 x 10$^{-3}$ |
| [11]      | a-SiGe / Planar  | 300       | 7.2 x 10$^6$           | n.a.             | 2.5 x 10$^9$              | 125 x 10$^{-3}$|
|           | a-SiGe / Trilayer| 300       | 2 x 10$^9$             | n.a.             | (1-40) x 10$^9$           | 0.1 x 10$^{-3}$|
| [12]      | a-SiGe / Planar  | 300       | 1 x 10$^6$ @ 250       | n.a.             | 6.7 x 10$^9$              | 13 x 10$^{-3}$ |
| [13]      | a-SiGe / Trilayer| 300       | 1.5 x 10$^6$ @ 10      | n.a.             | n.a.                      | 17 x 10$^{-3}$ |
|           |                  |           | 1 x 10$^6$ @ 200       | n.a.             | n.a.                      |               |
| [14]      | VWOx / Planar    | 300       | 5 x 10$^7$ @ 15        | 5.4 x 10$^{-10}$ | 1.1 x 10$^7$              | 7.2 x 10$^{-4}$ |
| [15]      | VWOx / Planar    | 300       | 6 x 10$^7$ @ 15        | 9 x 10$^8$       | n.a.                      |               |

$^a$ Measurement temperature; $^b$ Voltage responsivity at specified modulation frequency; $^c$ Noise equivalent power; $^d$ Detectivity; $^e$ Time constant; $^f$ Regular bolometric response; $^g$ Pyroelectric response.

4. Conclusion

Semiconducting a-YBCO is an attractive sensing oxide for IR thermal detection. a-YBCO thin films can be deposited on silicon substrates and thus allow integration with CMOS readout electronics; besides, the TCR compares favorably with other commercially used sensing materials. Planar and tri-layer
structures have been processed and tested. They both exhibit a high-pass pyroelectric response behavior, which was observed between 300 K and 95 K in the near IR; detectivity values up to $3.5 \times 10^8 \text{cm}^2\text{vHz/W}$ and 2-3 $\mu$s time constants have been measured. This resulting high frequency sensitivity offers a promising solution for fast imaging applications, especially in the far IR / THz range where moderate cost systems should be considered. Improvements are under progress concerning both the input radiation coupling with THz micro-antennas and the back-end electric coupling of these high impedance devices to correct the low-frequency response distortion and the low temperature response decrease.

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