Fast thermoelectric response of normal state $YBa_2Cu_3O_{7-\delta}$ films

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Normal state $YBa_2Cu_3O_{7-\delta}$ films, epitaxially grown "off-c axis" with tilt angles up to 20° are shown to be fast thermoelectric detectors for radiation from UV to far infrared wavelengths. Upon radiation heating of the tilted films a thermoelectric voltage arises due to the anisotropy of the thermopower in $YBa_2Cu_3O_{7-\delta}$. The response time is limited by the decaying temperature gradient and thus by heat diffusion. For thin films we have measured a response time from $\lesssim$1 ns in the UV to ~5 ns in the far infrared. Because of the wavelength dependent reflectivity, the responsivity of the films varies between 0.5 V/MW and 20 V/MW. Thus, thin tilted $YBa_2Cu_3O_{7-\delta}$ films can be used as fast room temperature detectors over a wide spectral range. © *1995 American Institute of Physics.*

Large amplitude voltaic response (~1 V) has been observed in normal state $YBa_2Cu_3O_{7-\delta}$ films (YBCO) subject to pulsed laser irradiation by several groups.[1–6] Recently it has been shown that these large signals arise due to a transverse thermoelectric effect which becomes effective in epitaxial films where the YBCO c axis is inclined by an angle α with respect to the macroscopic surface normal.[7] Films grown with a large tilt angle $\alpha \sim 10° - 20°$ show open circuit voltages up to 250 V for fluences slightly below the destruction limit. These films can be used as room temperature detector elements of high dynamic range, since their output voltage is proportional to the incident laser power over more than 11 orders of magnitude.[7] Up to now, investigations have been focused on relatively thick films ($d \geqslant 100$ nm) with response times $\tau \gtrsim 15$ ns. In this letter we report on an investigation of the response time and the responsivity of very thin films ($d = 35$ nm) over an extremely wide spectral range. Pulsed lasers ranging in wavelength from the UV to the far infrared (FIR) have been used to irradiate the tilted films.

Heating the surface of a thin film by the absorption of radiation establishes a temperature gradient ∇T perpendicular to the film surface. Due to the Seebeck effect a thermoelectric field

$$\mathbf{E} = S\ \nabla T \tag{1}$$

is generated, where the Seebeck tensor S shows an offdiagonal element[7]

$$S_{xz} = \Delta S\ \sin\alpha\ \cos\alpha \tag{2}$$

for our experimental configuration (see Fig. 1). $\Delta S = S_{ab} - S_c \sim -10\ \mu$V/K[8] is the difference of the absolute thermopowers in the ab plane and along the c axis of YBCO. S_{xz} gives rise to an electric field perpendicular to the temperature gradient leading to a voltage[7]

$$U = \Delta S \cdot \Delta T \cdot \frac{l}{d}\alpha, \tag{3}$$

where ΔT is the temperature difference between film surface and film bottom, l is the diameter of the illuminated spot, d the film thickness, and α has been assumed to be $\lesssim 20°$ so that the approximation $\sin\alpha\cos\alpha \approx \alpha$ holds. In comparison to a conventional thermocouple where a voltage $U_{\text{long}} = \Delta S\,\Delta T$ is obtained, the voltage in this transverse configuration is enhanced by a geometric factor $(l/d)\ \alpha$, which is of the order of $10^4 - 10^5$ for thin films with large tilt angles. For a 35 nm film with $\alpha = 20°$ and a laser spot with diameter $l = 5$ mm Eq. (3) gives a sensitivity of $U/\Delta T \sim 500$ mV/K. Therefore, even a small temperature difference induced by radiation heating leads to a substantial output voltage.

Epitaxial YBCO films with thicknesses between 35 and 500 nm were grown by standard laser ablation on $SrTiO_3$ substrates cut with the surface at an angle of $\alpha = 10°$ to the (100) plane. X-ray analyses showed that the films grow with the (100) planes parallel to the $SrTiO_3$ (100) planes, resulting in coherently tilted films with the c axis inclined by 10° to the surface normal. The films had T_c's between 84 K and 91 K, indicating good film growth. Electrical contacts were made from the films to 50 Ω coaxial cables with silver loaded conductive paint. Voltage signals were recorded with a 500 MHz bandwidth oscilloscope.

Figure 2 shows the response of a series of YBCO films with $\alpha = 10°$ to laser pulses from a XeCl excimer laser [$\lambda = 308$ nm, $t_p \approx 30$ ns (full width half-maximum)]. The 35 nm thick film almost reproduces the temporal structure of the laser pulse. The thicker films show an increasing response time, up to ~100 ns for the 400 nm film. Also shown in Fig. 2 with broken lines is the response of the films as calculated

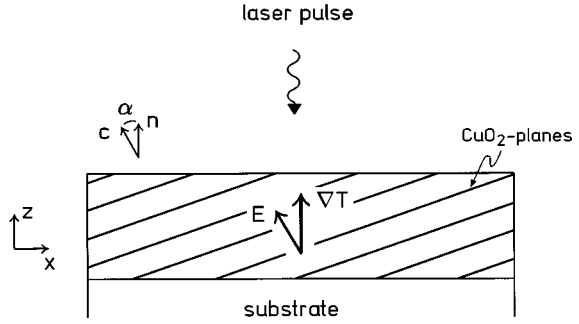


FIG. 1. Schematic cross section of a $YBa_2Cu_3O_{7-\delta}$ film, with the c axis tilted by an angle α with respect to the macroscopic surface normal n. The laser induced temperature gradient leads to a thermoelectric field **E** which has a component in the plane of the film.

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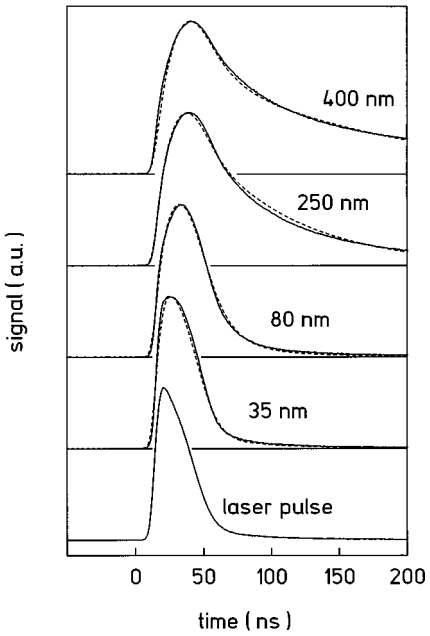


FIG. 2. Response of a series of tilted YBCO films to excimer laser pulses ($\lambda=308$ nm, $t_p\approx30$ ns). The bottom trace shows the laser pulse as recorded with a fast Si photodiode. The full lines show the measured response of the YBCO films, broken lines show the response calculated with a heat transfer model.

with a heat transfer model[9] using published data on thermal properties of YBCO and the YBCO–$SrTiO_3$ interface.[10] The close correspondence of measured and calculated traces indicates that such a thermal model is well suited to describe the time development of the thermoelectric voltage.

To investigate the response time for different wavelengths we selected the thinnest film ($d=35$ nm) for further study. The film was irradiated with laser pulses from a N_2 laser ($\lambda=337$ nm, $t_p\approx1$ ns), a dye laser ($\lambda=532$ nm, $t_p\approx5$ ns), a TEA CO_2 laser ($\lambda=10.6$ μm, $t_p\approx80$ ns), and an optically pumped FIR laser (CH_3F, $\lambda=495$ μm, $t_p\approx100$ ns with irregular pulse structure). Pulse energies varied between 10 μJ and 2 mJ. As reference detectors we used a fast pyroelectric detector (Molectron P5-01) for $\lambda=337$ nm, a vacuum photodiode (Hamamatsu S20) for $\lambda=532$ nm, a photon drag detector for $\lambda=10.6$ μm, and a fast intraband photoconductive Ge detector for $\lambda=495$ μm. Figure 3 shows the response of the film to these laser sources together with the response of the reference detectors, which have been inverted for clarity.

The fastest response is obtained at short wavelengths. At $\lambda=337$ nm the full width at half-maximum of the signal is 1.5 ns. The same width is found in the signal of the pyroelectric reference detector. The response time is therefore most likely limited by the duration of the laser pulse and the 500 MHz bandwidth of the oscilloscope. At larger wavelengths the response time increases up to ~5 ns in the FIR. This increase of the response time can be explained by considering the variation of the optical absorption depth. In the far infrared the absorption depth is several hundred nm,[11] leading to a relatively uniform heating of the film. The response time is then given by the time constant for heat loss out of the film[12]

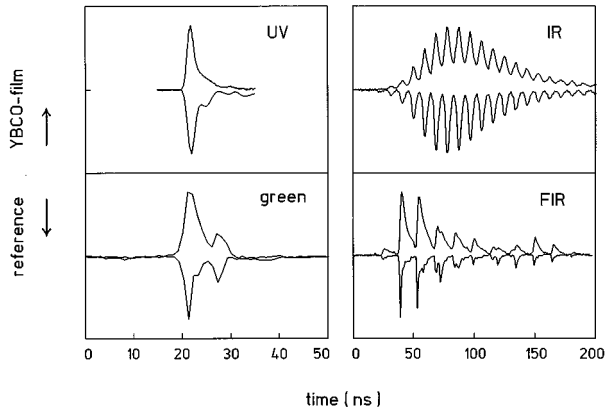


FIG. 3. Response of a 35 nm YBCO film to laser pulses at $\lambda=337$ nm, $\lambda=532$ nm, $\lambda=10.6$ μm, and $\lambda=495$ μm together with the response of different reference detectors. For clarity the response of the reference detectors has been inverted. The response time of the YBCO film ranges from $\leqslant1$ ns in the UV region to ~5 ns at far infrared wavelengths.

$$\tau_{\text{loss}}=C\,R_{\text{bd}}d, \tag{4}$$

where C is the heat capacity per unit volume of YBCO and R_{bd} is the thermal boundary resistance between film and substrate. Using $C=2.5$ J/cm^3 K and $R_{\text{bd}}=5\times10^{-4}$ K cm^2/W[10] Eq. (4) gives a response time of ≈5 ns for a 35 nm thick film, in good agreement with experiment. For UV wavelengths the absorption depth decreases to ~60 nm,[13] leading to a much steeper temperature distribution. Then the diffusion of heat through the film has to be taken into account, since this diffusion is effective in reducing the temperature gradient and hence the thermoelectric voltage. A characteristic time constant for heat diffusion within the film is[14]

$$\tau_{\text{diff}}=\frac{d^2}{2D}, \tag{5}$$

where D is the diffusivity of the film. With $D=7\times10^{-3}$ cm^2/s[10] one finds $\tau_{\text{diff}}\approx1$ ns for the 35 nm film, considerably smaller than τ_{loss}. Thus, the faster response at shorter wavelengths reflects the fact that due to a smaller absorption depth heat diffusion within the film rather than heat loss over the film–substrate boundary determines the decay of the temperature gradient.

To measure the responsivity of the films, a beam splitter diverted part of the laser pulse to a reference detector. Using a Gentec ED 200 pyroelectric joulemeter in the main beam, the reference detector could be calibrated to give the instantaneous laser power. Then the joulemeter was replaced by one of the YBCO films and the response of the film and of the reference detector was recorded simultaneously. This allowed to calculate the responsivity of the films. The results are displayed in Fig. 4 for three films with different thicknesses. The thinner films show a higher responsivity as is to be expected from Eq. (3). Maximum responsivity is obtained at short wavelengths (~20 V/MW) where the reflectivity of the films is only 10%–20%.[13] In the far infrared the reflec-

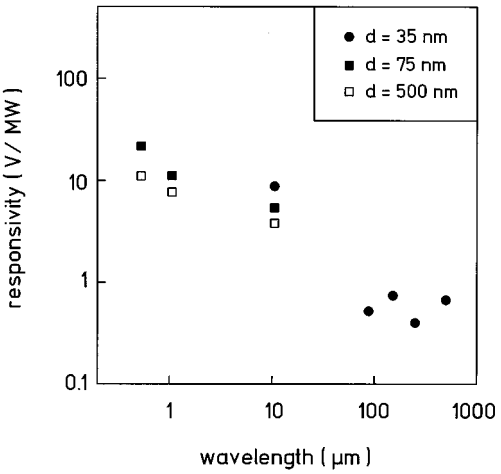


FIG. 4. Wavelength dependence of the responsivity of tilted YBCO films with different thicknesses. In the FIR the responsivity is low (0.5 V/MW) but relatively uniform between 80 μm and 500 μm. For shorter wavelengths the responsivity increases as the reflectivity of the films decreases.

tivity of the 35 nm film was measured to range from 90% to 95%, so most of the incident energy is reflected by the film. Accordingly the responsivity of (0.5 ± 0.2) V/MW is low, but relatively uniform between $\lambda=80$ μm and 500 μm.[15] These values of the responsivity are comparable to fast pyroelectric or photon drag detectors.

Equations (4) and (5) for the characteristic time constants suggest that by using still thinner films ($d\sim10$ nm) a response time of 100 ps can be achieved. Coherently tilted YBCO films might thus be the basis of thermal sub-ns room temperature detectors, combining high speed with the broad spectral sensitivity of thermal devices. We note that the tilted films are intrinsically faster than usual bolometric devices, since the output signal is proportional to the temperature gradient rather than to the average temperature increase of the film.

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