Normal state $YBa_2Cu_3O_x$ films: A new fast thermal detector for far infrared laser radiation with a uniform wavelength response

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Room temperature $YBa_2Cu_3O_x$ films, epitaxially grown on oriented substrates, are shown to be effective detectors of far infrared laser pulses. The photovoltage arises due to an anisotropic material thermopower subject to the thermal gradient arising from the radiation heating. For a 40 nm thick film we have measured a responsivity of $(5\pm2)\times10^{-7}$ V/W, independent of frequency in the range from 20 to 110 cm^{-1}. The response time is limited by thermal diffusion, giving a 200 MHz detector bandwidth and a noise equivalent power of 3.6×10^{-3} $W(Hz)^{-1/2}$ for this film thickness. Due to their large area (1 cm^2) and uniform spectral response, such films may be useful replacements for pyroelectric power/energy meters.

A frequent feature of room temperature power and energy meters for laser pulses is their strong wavelength dependence in the far infrared (FIR), i.e., for frequencies $20\ cm^{-1} < \omega < 100\ cm^{-1}$. Such strong wavelength dependences increase the difficulty of calibrating detectors and of making absolute and/or relative power measurements. The nonuniform spectral response may arise from the intrinsic detection mechanism, as is the case in photoconductive[1] and photon drag[2] devices, or from interference effects due to the detector crystal/electrodes, as has been observed in pyroelectric elements.[3,4] Obtaining a wavelength independent response therefore requires a frequency independent detection mechanism to be incorporated in a nonresonant detector structure. This communication suggests a new solution to this problem using thin films of the high temperature superconductor $YBa_2Cu_3O_x$ at room temperature as the detector element. Such films, when deposited on specially prepared substrates, have recently been shown to exhibit a thermoelectric photoresponse effective from UV to FIR wavelengths.[5,6]

The generation mechanism has been explained by the anisotropy of the thermopower in $YBa_2Cu_3O_x$.[5] In a plausible microscopic picture, different absolute thermopowers are ascribed to the metallic CuO planes and the more insulating intermediate layers. If these (100) planes are tilted by an angle α relative to the film surface (Fig. 1), they form a series connection of atomic scale thermocouples. Each consists of a pair of CuO planes with the intermediate material. Heating the film surface, for example by the absorption of radiation, establishes a temperature gradient within the film. Thus a thermoelectric voltage is generated. As this response is purely thermal, such films may be useful FIR detector elements.

Accordingly, we have studied the FIR response of several films. They were deposited by a standard excimer laser process on $SrTiO_3$ substrates cut with the surface normal at an angle of 10° to the (100) direction. The 1 cm^2 films were fixed in a metal box for electrical screening and electrical contacts were made from the film to 50 Ω coaxial cable with silver loaded conductive paint. Any signal obtained was amplified with a 50 Ω broadband low noise amplifier and recorded with a 500 MHz bandwidth oscilloscope. Our radiation source was a superradiant oversized metal waveguide device, optically pumped by a Lumonics 103 TEA CO_2 laser. The grating tuned pump laser has a multilongitudinal mode output, and the resulting irregular pulse structure is transferred to the FIR pulse. For the experiments that we will describe, FIR laser fill gases were CH_3F giving an output frequency of 20.2 cm^{-1} for a $9P(20)$ pump, 39.9 cm^{-1} from $9R(26)$, and NH_3 with output frequencies of 66 cm^{-1} from $9R(30)$ and 113 cm^{-1} from $9P(20)$ pump lines.[3]

FIR pulse profiles were obtained by diverting part of the beam with a Mylar beamsplitter into a fast (1 ns rise time) intraband photoconductive Ge detector.[7] Energies were measured with a Gentec ED200 pyroelectric joulemeter. This instrument, designed for visible and near infrared use, was found to have a reduced sensitivity in the FIR. This is because the black absorbing coating becomes increasingly transparent at long wavelengths, allowing the reflectivity of the pyroelectric crystal to affect the responsivity. Measurements made with a Fourier transform spectrometer and the FIR laser system described previously reveal the high, frequency dependent reflectivity (see Fig. 2). Here the Fourier spectrometer curve was scaled by a least squares procedure to best fit the laser data. To measure FIR pulse energies, it was assumed that the manufacturers' quoted responsivity was reduced by a factor of $(1-R)$, where R was the measured power reflectivity at the frequency of interest.

Figure 3 shows the response of a series of $YBa_2Cu_3O_x$ films to 40 cm^{-1} radiation. The bottom trace shows the signal from the photoconductive detector, while the next higher trace shows the response of a 40 nm film to the same pulse. It can be seen that the pulse substructure is reproduced, apart from the lengthened fall times of the peaks. From this decay time of 5 ns, we estimate the electrical bandwidth of the detector to be 200 MHz. Other traces in Fig. 3 show the response of thicker films to similar pulses: as the thickness increases the response time is reduced until, for a thickness of 300 nm, the pulse substructure is nearly indistinguishable from the background signal.

The response times of films with different thicknesses can be understood in terms of a thermal model.[8] In the

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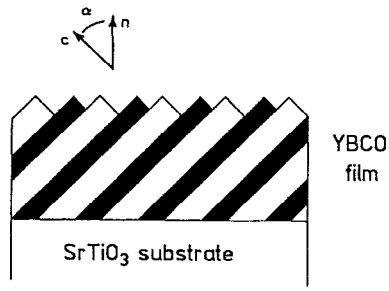

FIG. 1. Diagram showing the proposed arrangement of thermocouples within a $YBa_2Cu_3O_x$ film grown on an oriented $SrTiO_3$ substrate. Black and white areas represent layers of material with different absolute thermopowers, c denotes the direction of the film and substrate's c axis, which is inclined at an angle α to the surface normal n.

FIR, the Hagens–Rubens approximation is valid for $YBa_2Cu_3O_x$ at 300 K,[9] and this predicts an absorption depth scaling as $\omega^{1/2}$. Calculated values are 800 nm at 20.2 cm^{-1}, decreasing to 350 nm at 100 cm^{-1}. Thus, for all the films investigated, heating was relatively uniform, and the response times were determined by the heat flow across the film-substrate interface. This maintains the required thermal gradient across the film thickness.

Due to the faster response time, we selected the 40 nm film for further study. By using the energy meter in conjunction with the photoconductive detector and the pulse integration facility of the oscilloscope, instantaneous powers could be calculated at each frequency. The measured film responsivity as a function of frequency is displayed in Fig. 4. It indicates a relatively uniform sensitivity of $(5\pm2)\times10^{-7}$ V/W between 20 and 100 cm^{-1}. Such behavior is to be expected since (a) the film is much thinner than the radiation absorption depth and (b) neither $YBa_2Cu_3O_x$ (Ref. 9) nor $SrTiO_3$ (Ref. 10) have strong absorption features in this spectral region. Thus, suitably

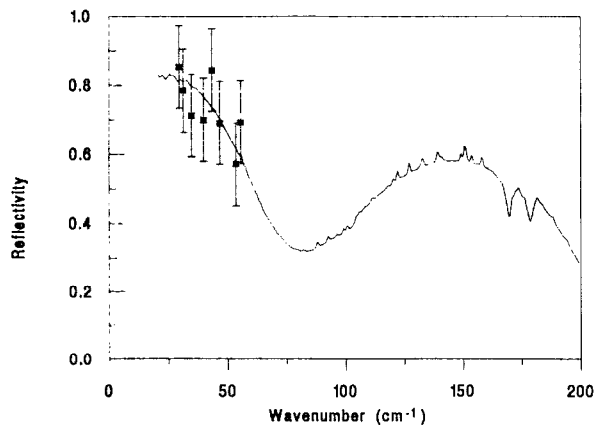

FIG. 2. Reflectivity of Gentec ED200 pyroelectric energy meter in the FIR. The solid line was obtained by a Fourier transform spectrometer with an instrument resolution of 0.5 cm^{-1}, points with error bars are the results of laser measurements. The strong variation in reflectivity with frequency is believed to arise from interference effects within the pyroelectric crystal and coating.

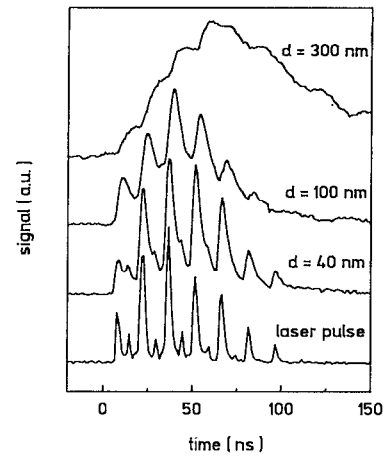

FIG. 3. Response of a series of $YBa_2Cu_3O_x$ films to 40 cm^{-1} laser pulses. The bottom trace shows the laser pulse as measured with a 500 MHz bandwidth photoconductive detector/oscilloscope combination. The signal from the 40 nm $YBa_2Cu_3O_x$ film is shown immediately above this. The responses of the two thicker films to similar laser pulses are also displayed.

prepared $YBa_2Cu_3O_x$ films exhibit a significant improvement in spectral response uniformity over conventional 300 K detectors of FIR laser pulses.

The linearity of the response was checked by the use of calibrated card attenuators: no significant deviations from linearity were found over the three orders of magnitude, even down to the noise limit imposed by the amplifier. In order to determine the detector noise, we used a low noise amplifier to observe the voltage fluctuations over a 3 Hz–2 MHz bandwidth, comparing the film with a metal film resistor of the same ohmic value. No difference could be discerned between the two sources. As the film operates without bias current, the noise equivalent power (N.E.P.) is thus determined by the Johnson noise as 3.6×10^{-3} $W(Hz)^{-1/2}$. Radiation induced fluctuations may also be a source of noise, but previous calculations show that their contribution would be negligible here.[11]

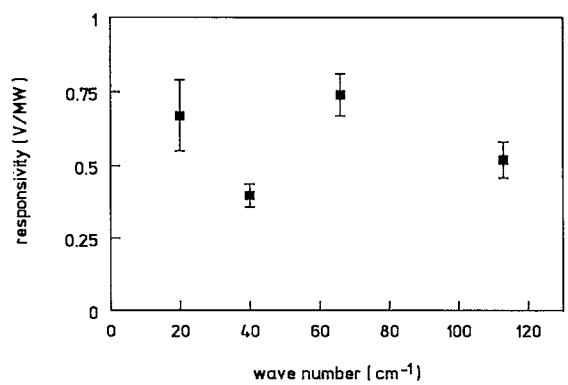

FIG. 4. Frequency dependence of the responsivity of the 40 nm $YBa_2Cu_3O_x$ film in a range from 20 to 113 cm^{-1}. The responsivity in this region is thus estimated as $(5\pm2)\times10^{-7}$ V/W. This relatively uniform response is in strong contrast to that of the pyroelectric detectors in Fig. 2 and other data (Refs. 3 and 4).

One of the drawbacks of such films as detectors is their relatively low responsivity. The FIR reflectivity of bulk $YBa_2Cu_3O_x$ at room temperature is over 90%[9] so most of the energy is reflected by the film. Considerable improvements might be achieved by any of several methods such as the use of a deposited absorbing coating,[12] incorporation of the film into an integrating cavity,[13] or by using multiple reflections from a series of films. However, care must be taken to avoid resonances when applying the latter two approaches, as these would compromise the spectral uniformity of the response.

In conclusion, we have shown that epitaxial $YBa_2Cu_3O_x$ films, deposited on correctly oriented substrates, are effective, large area, room temperature, solid state detectors of FIR laser pulses. In contrast to pyroelectric detectors, a relatively uniform FIR response has been measured at frequencies between 20.2 and 113 cm^{-1}. Using a 40 nm film as a prototype detector, we obtained a responsivity of $(5\pm2)\times10^{-7}$ V/W with a 200 MHz detector bandwidth and a N.E.P. of 3.6×10^{-3} $W(Hz)^{-1/2}$. Several suggestions of methods to increase the responsivity have been made.

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