Wavelength-dependent power-law Josephson photoresponse of a Tl₂Ba₂CaCu₂O₈ thin film

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The Josephson photoresponse of a current biased granular Tl₂Ba₂CaCu₂O₈ thin film has been measured at 4.2 K for radiation frequencies ω in the range 20-111 cm⁻¹. The resulting photosignal s depends nonlinearly on the incident power P satisfying the relationship $s = K(\omega)P^{\alpha(\omega)}$, where K is a frequency-dependent constant. The value of α increases monotonically from 0.5 at 20.2 cm⁻¹ to 0.9 at 111 cm⁻¹. At 20.2 cm⁻¹ this power law is valid for incident powers between 5 mW and 1 kW, a range of more than 105. These limits are imposed by experimental constraints and not by deviations from the power-law behavior. As the frequency increases, the responsivity of the film drops strongly, $K(\omega)$ decreasing approximately as ω^{-3} , so that the power-law validity is established over three orders of magnitude at 111 cm⁻¹. Such a behavior of the Josephson photoresponse has been previously observed, albeit over reduced dynamic ranges, in granular superconducting films of other materials. Thus we believe it to be an inherent characteristic of the random arrays of Josephson junctions contained in such films.

INTRODUCTION

The response of superconducting films to electromagnetic radiation has been widely studied: See, for example, Ref. 1 and references therein. However, much of this work has been concerned with the detection of visible and near-infrared light and the relevant photoresponse mechanisms. Much less attention has been given to detection in the far-infrared (FIR) and microwave region, i.e., the study of the response of films to photons with energies below or of the order of the superconducting energy gap. Granular films of both conventional^{2,3} and high-temperature superconductors⁴⁻⁹ have been shown to be effective radiation detectors for this spectral region. Such films behave as a random array of Josephson junctions, with the weak links occurring at the grain boundaries. Upon irradiation, the critical currents of the junctions are depressed. When the appropriate current bias is applied, a photosignal is thus developed across the film. As this photodetection process has a subnanosecond response time, effort has been invested to develop sensitive radiation detectors and to study the nature of the photoresponse process.

Initial work focused on microwave detection with Sn films, 2,10 but more recently NbN/BN has been proposed for the FIR region.³ First studies of detection with high-temperature superconductors took place on granular films of YBa₂Cu₃O_x, 4 since then further works have also dealt with this compound. 5,6 While the power and spectral responsivities of FIR detection using a Bi₂Sr₂CaCu₂O₈ film have been published, ^{7,8} relatively little work has taken place on thallium-based films. 9,11 Thus we have studied in detail the dependences of the Josephson photoresponse of a Tl₂Ba₂CaCu₂O₈ film on radiation frequency, incident power, and bias current. Measurements took place in the frequency range from 20.2 cm⁻¹ $< \omega < 111$ cm⁻¹, using a pulsed laser source to obtain incident powers P ranging from 1 mW to 1 kW.

The principal result of this work is that the photosignal

s is found to follow the power-law relationship

$$s = K(\omega)P^{\alpha(\omega)}, \tag{1}$$

where, for a given bias current I_b , the constant K and exponent α depend on the radiation frequency. Such a dependence was obeyed over several orders of magnitude in power, the limits being imposed by the range of the measurement technique. The value of the exponent α is found to increase with radiation frequency, rising from a value of approximately 0.5 at 20.2 cm⁻¹ to about 0.9 at 111 cm⁻¹. Furthermore the film sensitivity decreases strongly with increasing photon energy, so that it is only possible to measure over a range of 10³ in power at 111 cm⁻¹, compared to a factor of nearly 10⁶ at 20.2 cm⁻¹. At the measurement temperature of 4.2 K, the photosignal is found to depend approximately linearly on the bias current, independent of the laser wavelength.

Such a dependence, $s \propto P^{\alpha(\omega)}$, has previously been observed in measurements of the power dependences of FIR and microwave detection with granular films of other superconductor materials. 2,5,7,10 We therefore suggest that nonlinear detection, extending uniformly over several orders of magnitude, is a characteristic of Josephson photodetection with granular superconductor films. Furthermore, the value of the exponent α is found to depend strongly on the relative magnitudes of the photon energy and the superconducting energy gap 2Δ . For low photon energies, $h\nu \ll \Delta$, a signal proportional to the square root of laser power is in general observed. As the photon energy increases towards Δ , the exponent tends towards unity. In what follows we first describe the experimental technique. Experimental results pertaining to the Tl₂Ba₂CaCu₂O₈ film are then presented. In the discussion section, previous power dependence measurements on the far-infrared and microwave response of other granular superconducting films are briefly mentioned. It is shown that the dependences of other high- T_c compounds are in good agreement with the new results presented in this work. Finally, some of the implications of the observations as regards the photodetection mechanism are discussed.

EXPERIMENTAL

Measurements were made on a Tl₂Ba₂CaCu₂O₈ sample, formed by a process involving the diffusion of Tl into a precursor film deposited by excimer laser sputtering. 12 X-ray diffraction showed the 500-nm-thick film to be a single phase, with the c axis oriented normal to the MgO substrate. The $3 \times 2 \text{ mm}^2$ active area of the film was photolithographically patterned into an array of stripes, 13 each having dimensions of 60 μ m by 3 mm. Individual stripes were separated by a distance of 15 μ m. This patterning was designed so that the detector would behave as a transmission line with a characteristic impedance of approximately 50 Ω . The substrate was fixed to the cold finger of a liquid-helium cryostat and contacts were made to coaxial cables, as has been previously described. A four-point configuration was utilized, with separate 50- Ω lines for bias current and photosignal. Optical access into the cryostat was by a single TPX (poly-methylpentene) window.

The resistance-temperature characteristic is displayed as a semilogarithmic plot in Fig. 1. Upon cooling the film showed the typical resistance versus temperature curves of a granular film, ^{6,7,9} i.e., the main current independent decrease of resistance occurs at the bulk transition temperature due to the superconducting transition of the grains. At lower temperatures, current dependent tails can be observed in the characteristic; these arise from the intergranular Josephson junctions. Below about 40 K little change in the resistance with temperature was observable. Photoresponse measurements were thus made at 4.2 K so as to reduce the effects of any film heating due to absorption of the relatively high power incident radia-

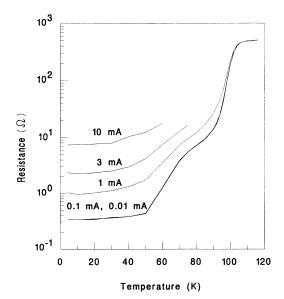


FIG. 1. The current dependence of the resistive transition of the $Tl_2Ba_2CaCu_2O_8$ film, shown on a semilogarithmic scale. The resistive tails at low currents are typical of granular high- T_c films: See Refs. 6, 7, and 9.

tion. Such heating would introduce a bolometric component into the photoresponse signal¹⁴ and would also thermally depress the critical currents of the junctions: Both effects are undesirable from the viewpoint of an investigation of Josephson detection and so should be minimized.

Our radiation source was a molecular gas laser, optically pumped by a transversely excited atmospheric (TEA) CO₂ laser. Reference 15 describes a similar system. Due to the electromagnetically noisy capacitor discharge required to pump the CO₂ molecules, it was necessary to place the laser in a shielded room to prevent detection of the associated radio frequency pulse by the film. The system produced irregular pulses of about 50-ns duration, with energies of up to 2 mJ, at repetition rates of 0.2 Hz. These FIR pulses displayed characteristic nanosecond scale fluctuations which arise from mode beating effects in the pump laser output. Thus signal averaging was not possible, and measurements were made on a pulse by pulse basis.

Due to the observed strong dependence of the superconducting films response on radiation frequency and also due to the wide range of powers employed, it was necessary to select FIR laser lines which were not accompanied by satellite emissions. ¹⁵ As our attenuators were less absorbing at low wave numbers, where the film is much more responsive, any lower frequency radiation component in the lasers output gave a falsely enhanced signal. Suitable monochromatic emission was found from FIR laser fill gases of CH₃F, with an emission frequency of 20.2 cm⁻¹, CO₂ laser pump line 9P(20); CH₃F, 40 cm⁻¹, 9R(26); NH₃, 66 cm⁻¹, 9P(34), and NH₃, 111 cm⁻¹, 9R(16).

Part of each laser pulse was diverted by a Mylar beam-splitter onto a fast intraband photoconductive Ge detector. The amplified detector output was recorded using a digital storage oscilloscope. We obtained the responsivity of the beamsplitter-detector combination at each frequency by using a calibrated pyroelectric Joule meter. ¹⁶ The remainder of the laser pulse was focused by an off-axis parabolic mirror to a diameter of approximately 8 mm at the sample position. The resulting photosignal was amplified with a 50- Ω impedance amplifier and stored with the same oscilloscope. Data points were obtained by comparing the peak signal heights from both detectors for the same laser pulse. The linearity of the Ge detector and the absorber attenuation was verified for each laser line by replacing the sample with the energy meter.

RESULTS

The dependence of the signal on the incident power at a frequency of 40 cm⁻¹ is presented in Fig. 2. For clarity, points corresponding to different bias currents have been scaled by the indicated factors. Each data point corresponds to an individual laser shot. The scatter amongst these is typical for measurements made with this type of laser system and it arises from the multispatial and temporal mode output radiation.

Within experimental error, it can be seen that the exponent of the power-law response does not depend on the bias current. For the largest applied current, 10 mA, this

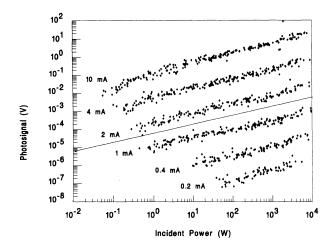


FIG. 2. The power dependence of the $Tl_2Ba_2CaCu_2O_8$ film photoresponse, at a frequency of 40 cm⁻¹, for the marked bias currents. Points corresponding to the different currents have been scaled for clarity as follows: 10 mA: \times 100, 4 mA: \times 10, 2 mA: \times 1, 1 mA: \times 0.1, 0.4 mA: \times 0.01, 0.2 mA: \times 0.001. The marked guide line indicates a slope of 0.5.

behavior is maintained over nearly five orders of magnitude in incident laser power. It is noteworthy that no significant deviation from the fixed exponent is observed at either very high or very low powers. By fitting the curves pertaining to different bias currents to Eq. (1), we calculate a value of $\alpha = 0.55 \pm 0.05$ at this frequency. The reduction in the measurement range at lower bias currents is imposed by noise, but even at the lowest applied current, $200 \mu A$, the dependence extends over about two orders of magnitude.

Similar power dependence experiments were performed for radiation frequencies of 20.2, 66, and 111 cm⁻¹. A current independent power-law response was observed at each wavelength accompanied by an approximately linear dependence of the signal on the bias current I_b . A fit to the experimental data gives a $s \propto (I_h)^{1.2 \pm 0.1}$ dependence when averaged over the four measurement frequencies. Two factors were observed to vary between the different data sets. Firstly the exponent increased with photon energy, as is illustrated in Fig. 3, which presents the dependence of the photosignal on incident power, at a fixed bias current of 2 mA, for the four measurement frequencies. Guidelines with slopes of 0.5 and 1 are included to show the change in α as ω increases. The value of α is found to increase monotonically from 0.49±0.06 at 20.2 cm⁻¹ to 0.90 ± 0.09 at 111 cm⁻¹. Additionally the responsivity of the film decreases strongly at higher frequencies. The effect of these factors is to curtail the measurement range at higher frequencies. Thus for a bias current of 10 mA, it was possible to measure the dependence over nearly six orders of magnitude at 20.2 cm⁻¹, but only over three orders at 111 cm⁻¹.

To extend measurements to lower powers, it is necessary to use chopped cw radiation and synchronous detection techniques. Using an optically pumped HCOOH gas laser with a radiation frequency of 23.1 cm⁻¹, incident powers of between 0.4 and 4 mW were achievable. In

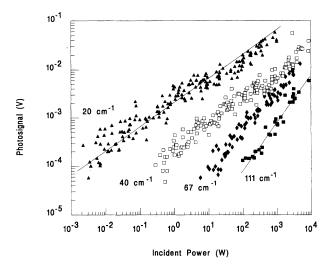


FIG. 3. The decrease of the $Tl_2Ba_2CaCu_2O_8$ film sensitivity and gradual transition from a $s \propto P^{0.5}$ regime to a $s \propto P^{0.93}$ behavior with increasing photon energy. Experimental points were obtained at a bias current of 2 mA. Radiation frequencies of the data sets are marked on the graph as are lines indicating slopes of 0.5 and 1.

this range a signal $\propto P$ was observed with a film sensitivity of about 1.2×10^{-3} V/W for a bias current of 1 mA. This Josephson signal was clearly distinguishable from a bolometric response observable at higher temperatures, because the amplitude of the former did not depend on the chopping frequency, in contrast to the thermal signal. ¹⁷

From the experimental data obtained with the pulsed laser source, it is possible to estimate the value of $K(\omega)$, the photosignal obtained for unit input power (1 W) at a given bias current. As the widest range of data is available at $I_b = 10$ mA, the dependence of $K(\omega)$ is plotted for this current in Fig. 4. It can be seen that $K(\omega)$ decreases strongly with ω : The line drawn on the graph indicates an arbitrarily chosen $1/\omega^3$ dependence. Such a plot is in-

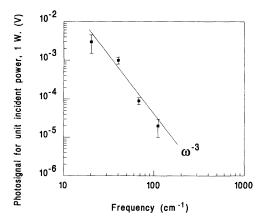


FIG. 4. Experimentally determined photosignals for unit incident power, 1 W, at the four measurement frequencies for a bias current of 10 mA. This gives the dependence of K, in units of V/W^{α} , on the photon energy. The line indicating an ω^{-3} dependence is intended as a guide to the eye only.

herently unreliable, because problems such laser beam spot size variations and Fabry-Perot effects cause a variation in the power incident on the film. However, a strong decrease in sensitivity with increasing ω has been previously reported for other granular films. ^{5,8} It is noted that this decrease in sensitivity may account for the failure to observe a Josephson photoresponse at 152 cm⁻¹ in a previous study of Tl₂Ba₂CaCu₂O₈. ¹¹ The decrease in $K(\omega)$ with ω would be difficult to analyze because of frequency dependent variations in (a) the coupling efficiency of the radiation to the antenna formed by the grid and (b) the conductivity of the film grains with the consequent variation in power coupled to the junctions and the skin depth.

DISCUSSION

As mentioned in the introduction, power-law responses have been previously observed by different groups working with both low- and high- T_c superconductors. In order to easily compare results from the microwave and FIR regions, it is convenient to introduce a normalized frequency Ω . This is defined as the ratio of the photon energy, $h\nu$, to the characteristic energy of a single junction: $\Omega \equiv h\nu/2eI_cR_n$, where I_c is the junction critical current and R_n its normal-state resistance. For high-quality, low capacitance tunnel junctions, at temperatures $T \ll T_c$ the quantity $2eI_cR_n$ reaches a maximum value of $\pi\Delta$. It is recognized that actual grain boundary junctions, especially in high- T_c materials often have a strongly reduced I_cR_n product.

Measurements of the microwave response of a granular Sn film, $\omega/2\pi = 9.4$ GHz corresponding to $\Omega \approx 0.05$, indicated a signal $\propto P^{0.5}$ over three orders of magnitude for bias currents varying by a factor of 25.2 The same group subsequently reported linear power response, $s \propto P$, over the same power range, at the approximately four times higher frequency of 38 GHz. 10 Unfortunately the measurement temperature was not given for the latter measurement, so the relevant value of Ω cannot be calculated. These measurements on the metallic superconductor are in excellent qualitative agreement with our observations on the Tl₂Ba₂CaCu₂O₈ film, namely the switch from an $s \propto P^{0.5}$ to an $s \propto P$ dependence as the photon energy increases, the uniform power dependence over several orders of magnitude, and the linear dependence of the signal on bias current.

Turning to the $\rm Bi_2Sr_2CaCu_2O_8$ system, we have previously reported 7 a $P^{0.5}$ dependence valid over three orders of magnitude in power at a film temperature of 17 K. With a laser frequency of 22.4 cm $^{-1}$, a lower limit to the value of Ω can be calculated by assuming a BCS-like energy gap and $T_c=85$ K: $\Omega_{\rm min}=0.07$. Using a granular film of nonstoichiometric YBa₂Cu₃O_x, a $P^{0.41}$ signal has been recently reported at 26 cm $^{-1}$, $\Omega_{\rm min}=0.1$, 5 for incident powers between 10 mW and 2 W at a sample temperature of 10 K. At power levels less than 10 mW, a sharp transition to a response $\propto P$ occurred. This transition has also been observed in the cw laser measurements on the $\rm Tl_2Ba_2CaCu_2O_8$ film, discussed above, and for the $\rm Bi_2Sr_2CaCu_2O_8$ film. 7 At the higher frequency of 152

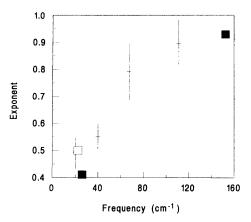


FIG. 5. Dependence of the exponent α on the laser frequency $(s \propto P^{\alpha})$. Values with error bars are obtained from the $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ film photoresponse, the solid squares are from measurements on a $\text{YBa}_2\text{Cu}_3\text{O}_x$ film (Ref. 5) and the open square represents $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ film data (Ref. 7).

cm⁻¹, $\Omega_{\rm min}$ =0.6, slightly sublinear detection, $s \propto P^{0.93}$, was also observed over a factor of 40 in power. Values of the exponent α obtained from the YBa₂Cu₃O_x film, and from the Bi₂Sr₂CaCu₂O₈ sample, are compared in Fig. 5 with the results from the Tl₂Ba₂CaCu₂O₈ film. This is reasonable as the differences in $\Omega_{\rm min}$ due to the variations in the bulk T_c of the different compounds, about 0.2 $\Omega_{\rm min}$, are probably insignificant compared with the variations in junctions within a given film and between films. It can be seen from Fig. 5 that the dependence of α on frequency for the Tl₂Ba₂CaCu₂O₈ film is in good agreement with the data from the earlier measurements on the other high- T_c compounds.

To the best of our knowledge, the only remaining measurements of the power dependence of the FIR photoresponse were made on granular film of NbN/BN.³ Here a signal $\propto P^{0.12}$ was found at 26 cm⁻¹, corresponding to $\Omega \approx 0.5$. For 87.7 cm⁻¹ radiation, $\Omega \approx 1.6$, and approximately $s \propto P$ behavior was measured. In particular, the low-frequency response is not in agreement with experimental results on Josephson detection with other film types. The authors have drawn attention to this anomalously low α value, and they suggest that a different photoresponse mechanism operates in this very thin (10 nm) film. This may be radiation induced vortex-antivortex depairing, which is expected to be effective in such a quasi-two-dimensional system. 20 The presence of such depairing is indicated by a power-law dependence of resistance on bias current. 20 Thus this photodetection process may be excluded as regards the Tl₂Ba₂CaCu₂O₈ film, on account of both the 500-nm film thickness and the approximately linear resistance-current characteristic for currents greater than 0.1 mA.

To summarize, for $\Omega \ll 1$, a photosignal proportional to $P^{0.5}$ over several orders of magnitude, accompanied by a linear response at lower powers, is an experimental characteristic of pure Josephson detection in thin films. As Ω tends towards unity, the exponent increases and the film sensitivity decreases sharply. To deal with this theoretically, it is necessary to discuss the photoresponse

of a single junction as we are not aware of applicable calculations for the response of a film. A percolative description has been applied to the transport and microwave detection characteristics of thin granular YBa₂Cu₃O_x films, assuming an exponential distribution of junction critical currents. 6,21 Unfortunately this model assumes a temperature close to T_c , and so it is not applicable to this study. We are not aware of any other photoresponse calculations. Film characteristics have also been previously explained in terms of a single effective junction, 22 but such an approach is not appropriate here due to the wide range of the power-law response.

For values of $\Omega < 1$, the impedance of a single junction is dominated by its resistance, 23 and it is therefore current biased at the radiation frequency. Calculations, based on the resistively shunted junction model, have shown that the critical current, I_{c0} , of such a junction is depressed by below-gap radiation. 24,25 Thus a photosignal results if the junction is current biased in the vicinity of its critical current. 26 For an impressed alternating current amplitude $I_{\omega} \ll I_{c0}$, the depression of I_c is proportional to I_{ω}^2 and hence to the radiation power P. For larger values of I_{ω} , approximately in the range $\Omega I_{c0} < I_{\omega} < I_{c0} (1+\Omega),^{25}$ the depression is proportional to I_{ω} and hence to $P^{0.5}$. Any further increase in I_{ω} results in an oscillatory behavior of I_c . These calculations have been verified by measurements on single junctions. 23

This for a value $\Omega=0.1$, $s \propto P^{0.5}$ is predicted over a factor of 10 in induced current or a range of 10^2 in incident power. At higher values of Ω , the relative size of the region of parabolic depression grows at the expense of the linear part, and so a reduced range of $P^{0.5}$ detection is expected. For our film $\omega=20.2$ cm⁻¹ corresponds to $\Omega_{\min}=0.07$. Thus a photosignal $s \propto P^{0.5}$ is expected at maximum over a range of 200 in power. This falls far short of the measured range of about 10^6 , so that in this simple picture junctions with a variation of approximately 10^3 in critical current are responsible for the detection. A further problem with the resistively shunted junction model is that it predicts only $s \propto P^{0.5}$ and $s \propto P$ detection and not the experimentally observed continuous variation

in α . Any theoretical approach must take into account the coupling of the junctions in the film to give intermediate and constant values of α over a wide range of powers. One problem facing such an analysis is the unknown character and location of the junctions responsible for the radiation detection. Some progress has recently been made towards solving this problem, when Shapiro steps were observable in a patterned granular film of YBa₂Cu₃O_x at a radiation frequency of 19.5 cm⁻¹. ²⁷ The manufactured constriction was of the order of the grain size, so that only a few Josephson junctions were responsible for the *I-V* characteristic.

CONCLUSION

It has been established that nonlinear photodetection, $s \propto P^{\alpha}$, 0.5 < α < 1, is a common feature of the FIR and microwave response of granular superconducting films. The value of α is found to increase monotonically with photon energy, but conclusions cannot be drawn about its ω dependence due to our lack of knowledge about the set of junctions responsible for the photoresponse of a film. For the compound $Tl_2Ba_2CaCu_2O_8$, values of α were found to be independent of laser powers varying by up to a factor of 10⁶. Saturation effects were not observed, even at a relatively high maximum incident powers of about 1 kW. Measurements made on other high- T_c thin films provide values of α in agreement with those measured in this study, α increasing from about 0.5 at 20 cm⁻¹ to approximately unity at 110 cm⁻¹. Furthermore, all films studied showed a strong decrease in sensitivity as the photon energy decreased. Applications of this P^{α} detection for the reception of radiation in the FIR range are not obvious. Advantages may lie in the increased film sensitivity at low frequencies, the unique noise properties of \sqrt{P} detection²⁸ and the very wide dynamic range ob-

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