

# Enhancement of Josephson photoresponse of granular high- $T_c$ superconductor thin films by deoxygenation

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The dependence of the far infrared Josephson photoresponse of current biased granular high- $T_c$  superconductor thin films on deoxygenation is presented.  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  and  $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  thin films were heated in vacuum to temperatures of between 200 and 500 °C. The resulting deoxygenation weakens the intergrain coupling, thereby reducing the critical current and enhancing the photoresponse. In this way the optimum temperature for the fast Josephson response may be tuned to lie outside the temperature region of the slow bolometric signal.

Fast nonbolometric photoresponse mechanisms in high- $T_c$  superconductors have recently attracted much attention.<sup>1–6</sup> Generally, nonequilibrium electronic effects have been observed in the visible to near infrared from epitaxial films, whereas in granular films the photoresponse originates from grain decoupling due to the local weakening of superconductivity at the boundaries.<sup>3</sup> Combinations of the two effects may also be observable.<sup>7</sup> The resistive state due to grain decoupling has been explained in terms of a Kosterlitz–Thouless phase transition,<sup>8</sup> vortex creation,<sup>9</sup> or the Josephson effect.<sup>10</sup> The far infrared (FIR) Josephson photoresponse is based on the latter effect,<sup>11,12</sup> utilizing the depression of the critical current  $j_{c0}$  of a junction by the radiation induced high frequency currents  $I_{\omega}$ . Photoresponse times, due to the above effects, are measured or estimated to be in the picosecond range.

In the FIR the quantum energy of the radiation is smaller than the BCS energy gap. Therefore, no pair breaking is expected for temperatures  $T \ll T_c$  and any nonbolometric response arises from weak links. Such an effect is very promising for the development of extremely fast FIR detectors based on granular films. These can easily be patterned to act as distributed antennas with signals arising from a superposition of all grain boundaries, or can be incorporated into antenna structures to achieve good radiation coupling, as it has been shown for bolometers.<sup>13</sup>

Nonbolometric microwave detection in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  has been linked to the material's granularity,<sup>14</sup> but no systematic studies of film production techniques to optimize the material's FIR response are known to us. The response studies generally focus on samples with poor superconducting behavior such as low critical currents and broad resistive transitions, whose reproducible production is difficult. The aim for optimizing Josephson detection has to be to produce films containing high quality grains, which are only weakly Josephson coupled. In classical superconductors this has been achieved, for instance, by embedding Nb in a BN matrix.<sup>15</sup> In this letter we present the influence of a vacuum annealing procedure on high quality films, which can reliably and reproducibly be manufactured and

we show that this treatment improves the detection characteristics in the way suggested above.

The studies were made on 300-nm-thick  $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  thin films on MgO substrates, mechanically patterned into stripes 50–500  $\mu\text{m}$  wide and 2 mm long. They were produced by standard pulsed excimer laser deposition and *ex situ* annealing which yielded granular *c*-axis oriented films with rocking curves of less than 0.2° [full width at half-maximum (FWHM), 0010 peak] and grain sizes of 1–5  $\mu\text{m}$ . Critical currents exceeding  $10^4$  A/cm<sup>2</sup> have been achieved at  $T < 25$  K. The stripes were constant current biased and the voltage response to 80 ns FIR pulses of about 1 W/cm<sup>2</sup> power densities at  $\lambda = 447$   $\mu\text{m}$  was observed, as described previously.<sup>11,12</sup>

Figures 1 and 2 show, respectively, the resistance and photoresponse curves of an untreated  $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  sample. In the photoresponse plot two superimposed maxima can be identified: One corresponds to the bolometric response at  $T \approx 105$  K, the other, peaking at  $T_p \approx 95$  K, is due to the Josephson response. For a useful high speed detector application these maxima are too close: Any signal here will also show a slow bolometric contribution. A shift of the Josephson peak to lower temperatures can be obtained by the post annealing procedure described below.

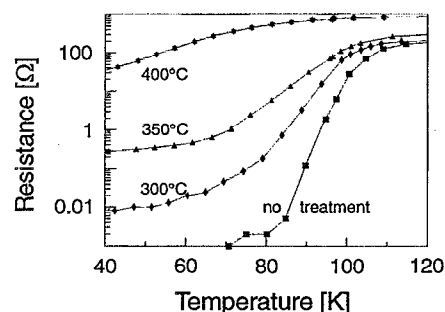


FIG. 1. Resistance vs temperature curve of a 250- $\mu\text{m}$ -wide  $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  stripe, for a bias current density of 1 kA/cm<sup>2</sup>, untreated and after subsequent vacuum heat treatments at increasing temperatures.

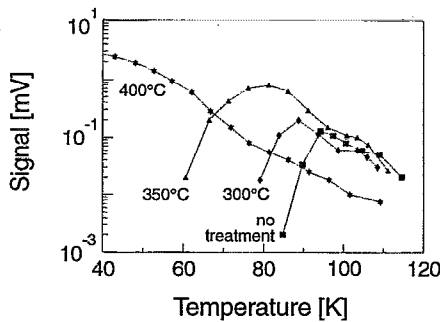


FIG. 2. Photoresponse vs temperature of a 250- $\mu\text{m}$ -wide  $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  stripe, for a bias current density of 1  $\text{kA}/\text{cm}^2$ , untreated and after subsequent vacuum heat treatments at increasing temperatures.

Following the  $R(T)$  and photoresponse measurement the silver paint contacts were removed and the samples cleaned with acetone. They were then placed in a quartz tube which was evacuated to a pressure  $p < 10^{-3}$  mbar and put into a tube furnace. The oven temperature was increased at a rate of 10 K/min from room temperature to the set temperature  $T_T$  and kept constant there for 30 min. After this treatment the samples were cooled rapidly under vacuum to room temperature. Then the resistance and photoresponse of the samples were measured again and the process was repeated on the same samples at an increased deoxygenating temperature. Values of  $T_T$  between 200  $^\circ\text{C}$  and 500  $^\circ\text{C}$  have been investigated.

The changes in the resistance are shown in Fig. 1. It can be seen that the vacuum treatment increases the film granularity: The normal state resistance, the midpoint, and the width of the transition are only slightly influenced for moderate heat treatments ( $T_T < 350$   $^\circ\text{C}$  for  $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  and  $T_T < 250$   $^\circ\text{C}$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ ). However,  $j_{c0}$  is depressed considerably and a resistance tail develops at low temperatures. This indicates that the bulk properties of the grains have not been changed and that only the grain boundaries are affected. At higher annealing temperatures the bulk material itself suffers a degradation and finally loses its superconducting properties.

We attribute this behavior to oxygen loss, as has been observed in granular bulk material:<sup>16</sup> At moderate temperatures oxygen will be removed from the grain surface and the boundaries only. Due to the reduced carrier concentration this creates a layer of reduced order parameter around the crystallites, reducing the intergrain coupling and thus explaining the  $j_{c0}$  reduction and the tail in the  $R(T)$  curve. Similarly, a higher normal state boundary resistance is caused, leading to the slight increase in the film's normal state resistance.

As can be seen from Fig. 2, the Josephson photoresponse increases and shifts to lower temperatures with increasing  $T_T$ , as can be expected from the reduced Josephson coupling seen in the resistivity measurements. This is clarified in Fig. 3:  $T_p$  decreases both with increasing  $T_T$  and  $j_b$ , so that  $T_p$  always coincides with the onset of a substantial resistance ( $R \approx 10$   $\Omega$  here). This is the typical behavior of the Josephson photoresponse:<sup>11,12</sup> the critical

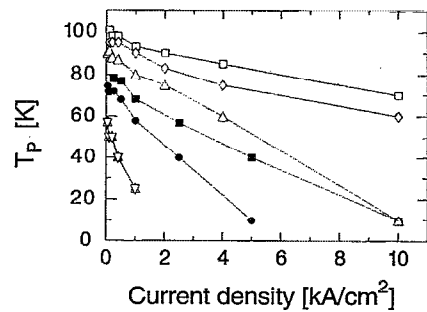


FIG. 3. Bias current dependence of the temperature of maximum Josephson photoresponse ( $T_p$ ) following different heat treatments. Open symbols are for  $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  film, filled symbols for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  film. The symbols correspond to the following vacuum annealing temperatures: No annealing (squares), 200  $^\circ\text{C}$  (circles), 300  $^\circ\text{C}$  (diamonds), 350  $^\circ\text{C}$  (triangles), and 400  $^\circ\text{C}$  (stars).

current density  $j_{c0}$  is reduced to a value  $j_c(I_w)$  by the FIR radiation and a voltage develops across those junctions which are biased with a current  $j_b > j_c(I_w)$ . Peak response occurs when a maximum number of junctions is biased at the value of their critical current density  $j_b = j_{c0}$ . The maximum of the photoresponse is therefore a measure for the  $j_{c0}$  of the “typical” junction in the current path. This is obviously higher than the  $j_{c0}$  as determined from transport current experiments which measure the weakest link. It also explains why the maximum response occurs in the partially resistive state and not right at the onset of resistivity. Therefore, Fig. 3 can also be interpreted in a different way: The temperature dependence of the critical current of a typical junction in the current path is displayed. For all treatments this behavior does not follow the Ambegaokar–Baratoff dependence for tunnel junctions, but shows more the behavior of SNS or flux flow dominated resistivity in granular films.<sup>17</sup> Thus, we propose that the nature of the Josephson decoupling of the grains in the films is due to fluxon movement across the junctions. This provides a microscopic explanation of previous macroscopic approaches to the FIR and mm-wave photoresponse<sup>8,14</sup> based on the decrease of the Josephson coupling energy with  $j_{c0}$  compared to the thermal energy. Our model also incorporates naturally the bias current dependence of the response maximum.

Figure 4 shows the bias current dependence of the peak photoresponse after treatments at different temperatures. Generally, a linear dependence of the photoresponse on bias current is found. Vacuum annealing shifts the response curves to higher signal values. However, for films treated at 250  $^\circ\text{C}$  ( $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ ) and 400  $^\circ\text{C}$  ( $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$ ) a saturation is seen. This occurs when the bias current exceeds the critical current of the typical grain boundary even at the lowest measurement temperatures. Also included is the effect of reoxygenating the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  sample in air for 30 min at 520  $^\circ\text{C}$ : The properties of the sample after the 200  $^\circ\text{C}$  treatment are restored.

Vacuum annealing improves the applicability of high quality granular films as fast FIR detectors due to two facts: The bias current needed for an optimum response at

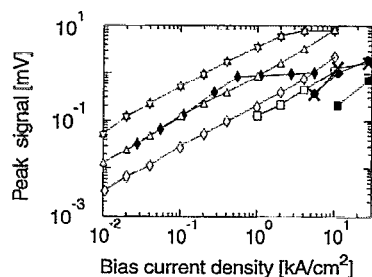


FIG. 4. Bias current dependence of the peak photoresponse with the temperature adjusted to  $T = T_p(j_b)$  for a 250- $\mu\text{m}$ -wide  $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$  stripe (open symbols) and a 100- $\mu\text{m}$ -wide  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  stripe (filled symbols) after subsequent annealing treatments: No annealing (squares), 200 °C (circles), 300 °C (diamonds), 350 °C (triangles), and 400 °C (stars) in vacuum and reannealing at 520 °C in air (crosses).

a given temperature is reduced, so that contact heating and noise is reduced and currents are more compatible with low noise high speed electronics. Further, the Josephson photoresponse at these moderate bias currents (of the order of 1 mA) is shifted to temperatures well below the midpoint of the transition and thus no slow bolometric contribution to the response occurs. For fixed operating temperature and bias current the films can be heat treated to show optimum performance.

By the annealing process we have achieved a standard sensitivity of  $0.1\text{V}/\sqrt{\text{W}}$  (the signal is proportional to  $\sqrt{\text{power}}$ ),<sup>11</sup> and a noise equivalent power of  $10^{-16}\text{W/Hz}$  using a simple stripe geometry. For optimum response bias current densities of  $10^3\text{A/cm}^2$  are sufficient, the detection at different operating temperatures can be optimized simply by adjusting the bias current and a clear separation of the fast Josephson photoresponse signal from the slow bolometric response is possible for temperatures  $T \lesssim T_c$ . Starting with high quality films a good reproducibility of the results is achieved. Additionally, we have shown that even moderate heating ( $T > 200\text{ °C}$ ) of superconducting

films strongly degrades the superconducting properties by enhancing the granularity and increasing the surface resistance<sup>18</sup> which is important for many applications.

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- <sup>1</sup>Gi. Schneider, H. Lengfellner, J. Betz, K. F. Renk, and W. Prettl, *Int. J. Infrared & Millimeter Waves* **12**, 1 (1991).
- <sup>2</sup>U. Strom, J. C. Culbertson, S. A. Wolf, F. Gao, D. B. Tanner, and G. L. Carr, *Phys. Rev. B* **46**, 8472 (1992).
- <sup>3</sup>M. Leung, P. R. Broussard, J. H. Claassen, M. Osofsky, S. A. Wolf, and U. Strom, *Appl. Phys. Lett.* **51**, 2046 (1987).
- <sup>4</sup>A. D. Semenov, G. N. Gol'tsman, I. G. Gogidze, A. V. Sergeev, P. T. Lang, and K. F. Renk, *Appl. Phys. Lett.* **60**, 903 (1992).
- <sup>5</sup>M. Johnson, *Phys. Rev. Lett.* **67**, 374 (1991).
- <sup>6</sup>L. Shi, G. L. Huang, C. Lehane, J. P. Zheng, and H. S. Kwok, *Appl. Phys. Lett.* **61**, 489 (1992).
- <sup>7</sup>K. Tanabe, Y. Enomoto, and A. Yamaji, *Appl. Phys. Lett.* **57**, 2719 (1990).
- <sup>8</sup>J. C. Culbertson, U. Strom, S. A. Wolf, P. Skeath, E. J. West, and W. K. Burns, *Phys. Rev. B* **39**, 12359 (1989).
- <sup>9</sup>A. M. Kadin, M. Leung, A. D. Smith, and J. M. Murduck, *Appl. Phys. Lett.* **57**, 2847 (1990).
- <sup>10</sup>M. A. Dubson, S. T. Herbert, J. J. Calabrese, D. C. Harris, B. R. Patton, and J. C. Garland, *Phys. Rev. Lett.* **60**, 1061 (1988).
- <sup>11</sup>P. G. Huggard, Gi. Schneider, T. O'Brien, P. Lemoine, W. Blau, and W. Prettl, *Appl. Phys. Lett.* **58**, 2549 (1991).
- <sup>12</sup>Gi. Schneider, P. G. Huggard, T. O'Brien, P. Lemoine, W. Blau, and W. Prettl, *Appl. Phys. Lett.* **60**, 648 (1992).
- <sup>13</sup>M. Nahum, Qing Hu, P. L. Richards, S. A. Sachtjen, N. Newman, and B. F. Cole, *IEEE Trans. Magn.* **27**, 3018 (1991).
- <sup>14</sup>A. S. Afanasyev, A. V. Volkov, V. N. Gubankov, Yu. Ya. Divin, and P. M. Shadrin, *IEEE Trans. Magn.* **25**, 2571 (1989).
- <sup>15</sup>M. Leung, U. Strom, J. C. Culbertson, J. H. Claassen, S. A. Wolf, and R. W. Simon, *Appl. Phys. Lett.* **50**, 1691 (1987).
- <sup>16</sup>K. Donnelly, J. F. Lawler, J. M. D. Coey, and B. Raveau, *Supercond. Sci. Technol.* **4**, 27 (1991).
- <sup>17</sup>J. W. C. De Vries, G. M. Stollman, and M. A. M. Gijs, *Physica C* **157**, 406 (1989).
- <sup>18</sup>D. Miller, P. L. Richards, S. Etemad, A. Inam, T. Venkatesan, B. Dutta, X. D. Wu, C. B. Eom, T. H. Geballe, N. Newman, and B. F. Cole, *Appl. Phys. Lett.* **59**, 2326 (1991).