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Superconducting Energy Gap from Infrared Response Measurements in Granular $Tl_2Ba_2CaCu_2O_8$ Films.

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Abstract. – The resistance response of polycrystalline $Tl_2Ba_2CaCu_2O_8$ films to short laser pulses has been investigated for a wide range of far-infrared frequencies. For temperatures $T \ll T_c$ a drastic change in response time is observed at a critical frequency $\nu_c \simeq 4$ THz. The time constant increases from about 1 ns at $\nu < \nu_c$ to 1 μs at $\nu > \nu_c$. The critical frequency is attributed to a superconducting energy gap with $2\Delta(0)/k_B T_c \simeq 2$.

A central problem of the physics of high-T_c superconductors is the determination of the superconducting energy gap. Several spectroscopic techniques have been applied, namely single-particle tunnelling [1], far-infrared [2, 3], Raman- [4], and photoelectron [5] spectroscopy. In this letter we report on a novel method based on the resistance response of granular films to short far-infrared (FIR) laser pulses. In previous investigations on superconducting films operated in a current biased mode, both slow bolometric signals and high-speed nonthermal response have been observed [6-13]. In our study we analysed the optical response of granular $Tl_2Ba_2CaCu_2O_8$ films in a wide range of FIR frequencies and found that the fast response occurs only below a critical frequency. Above this frequency only slow bolometric signals are observed for all temperatures below T_c. We associate the critical frequency with the energy gap of the superconductor.

Granular $Tl_2Ba_2CaCu_2O_8$ films were deposited on (100) $SrTiO_3$ and MgO substrates by a procedure minimizing thallium contamination of the growth set-up. At first thallium-free films were prepared by excimer laser ablation from a Ba-Ca-Cu-O ceramic of appropriate composition. Then the films were heated in contact with a $Tl_2Ba_2CaCu_2O_8$ ceramic sample yielding incorporation of thallium by diffusion. A final annealing step generated an almost homogeneous thallium distribution in the films. The resulting films were polycrystalline with random orientation of grains. Details of the film preparation and characterization are described elsewhere [14]. The films showed an onset of resistive current flow at about 40 K for vanishingly small current, decreasing with rising bias current. Strong increase of the resistivity occurred up to $T_c \simeq 100$ K, above this temperature the resistivity increased almost linearly with temperature.

Measurements were carried out on bridgelike structures of 1 μm thickness and (1×5) mm^2 area. Electrical contacts were prepared by silver epoxy and the samples were placed in a temperature-variable cryostat with optical access. The films were biased in series with a 50 Ω load resistor using a constant current source. In order to prevent irradiation of the contacts, the contact regions were shielded by a metallic mask. The signal voltage across the superconductor due to single laser pulses was recorded by a digital storage oscilloscope. A multimode TEA-CO_2 laser emitting pulse trains due to self-mode-locking and a FIR molecular (CH_3F and D_2O) laser pumped by the TEA-CO_2 laser were used covering a wavelength range between 10 μm and 496 μm. The infrared and FIR radiation was monitored by a broad-band GaAs Schottky diode. The mode beating structure was also present in the FIR emission.

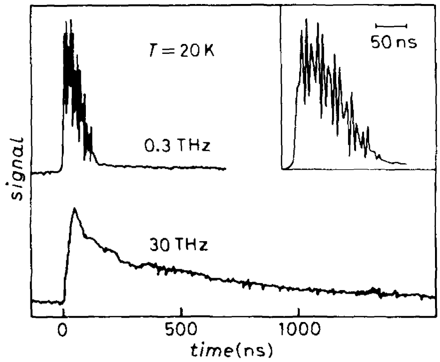

Fig. 1. – Optical response of a granular $Tl_2Ba_2CaCu_2O_8$ film below and above the critical frequency $\nu_c \simeq 4$ THz. Upper curve and inset: $\nu = 0.3$ THz; lower curve: $\nu = 30$ THz.

Measurements for two different photon energies at $T \simeq 20$ K are shown in fig. 1. For low frequency (upper curves) the optical response reproduces the temporal structure of the laser pulse within the limit of the bandwidth of the recording device. The rapid modulation of the signal is more clearly seen on the extended time scale in the inset of fig. 1. Our results demonstrate fast optical response with an upper limit of about 1 ns for the response time. On the other hand, radiation at high frequencies leads to a drastically slower response as shown by the lower trace in fig. 1. The signal decreases nonexponentially in a time of about 1 μs (10 per cent of peak value). The observed signal is characteristic of a bolometric response due to heating of the sample. The temporal structure of the signal pulse is determined by heat diffusion within the superconducting film and from the film to the substrate.

The response time as a function of radiation frequency is shown in fig. 2 for a temperature T ($\simeq 20$ K) far below T_c. Up to a critical frequency $\nu_c \simeq 4$ THz the response is fast. Near this frequency the time constant increases within a small frequency interval by three orders of magnitude. We attribute this critical frequency to a superconducting energy gap and find $2\Delta(0)/k_B T_c \simeq 2$.

As shown in fig. 3 for a frequency $\nu \simeq 0.6$ THz ($\lambda = 496$ μm), the fast signal is only observable at temperatures below T_c. At low temperature (upper curve) only the fast response is present while for a temperature (80 K) near T_c a small bolometric signal arises in addition to the fast response. Just above T_c (lower curve) only a bolometric response is

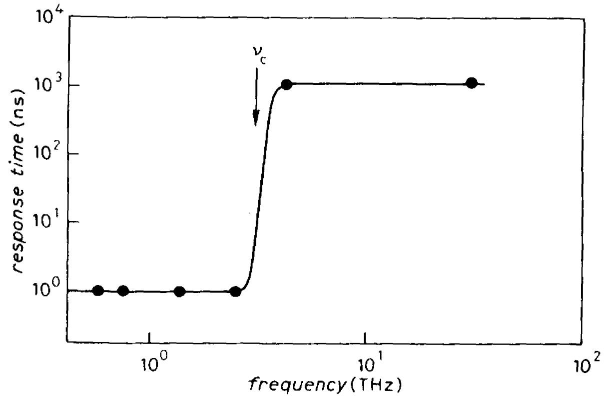

Fig. 2.

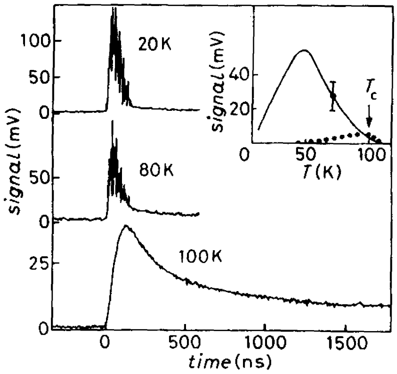

Fig. 3.

Fig. 2. - Response time *vs.* frequency at 20 K.

Fig. 3. - Optical response below ν_c ($\nu = 0.6$ THz) for various temperatures. Inset: signal height of the fast (line) and slow (points) signal *vs.* temperature, at a bias current of 10 mA and a laser pulse energy of $\sim 2\,\mu J$.

observed. The inset of fig. 3 shows the temperature dependence of the two signals. The fast signal attains a maximum at about 50 K and vanishes near T_c, while the bolometric signal has a maximum in the region around T_c. The decrease of the fast signal below 50 K towards lower temperatures was different for different samples and depended on the bias current. Our results clearly demonstrate that the fast signal is a property of the superconducting state.

In the superconducting state the current-carrying film represents a random array of grains coupled by weak links or Josephson junctions. Infrared irradiation induces a current that adds to the d.c. bias current. If the total current exceeds the critical current, a voltage response arises. Connected with this process there is energy dissipation at grain boundaries produced by relaxation and recombination of quasi-particles. The corresponding heat is rapidly removed to the superconducting grains because of the small dimensions of grain boundaries (some nm). This leads to the fast optical response for frequencies smaller than the critical frequency. If the photon energy is larger than the energy gap, light absorption can occur within the grains heating a substantial part of the film. This causes the bolometric signal with a response time in the order of $d^2/2k$ where $d \simeq 1\,\mu m$ is the film thickness and $k \simeq 0.01\,cm^2/s$ the thermal diffusivity [15]; a similar value for the thermal diffusivity has been obtained from the Nernst effect induced by laser heating observed for the same $Tl_2Ba_2CaCu_2O_8$ film [16]. The bolometric signal peaks where the derivative dR/dT has its maximum, namely around T_c.

We found that the fast signal saturated for our samples at a power density of the order of $5\,kW/mm^2$ and at a current density of the order of $50\,A/mm^2$. These observations are consistent with radiation-induced currents at grain boundaries. Saturation is reached if almost all grain boundaries contribute to the signal.

We note here that also in experiments with thick granular $YBa_2Cu_3O_{7-\delta}$ films fast response has been found in the FIR spectral region [7]. Slow response from granular $YBa_2Cu_3O_{7-\delta}$ films was obtained in the near infrared (for $1\,\mu m$-radiation) [8] and also from epitaxial $YBa_2Cu_3O_{7-\delta}$ films using infrared ($10.6\,\mu m$) and optical ($0.63\,\mu m$) radiation [9]. In a recent experiment with pulsed synchrotron radiation, bolometric response was obtained from epitaxial $YBa_2Cu_3O_{7-\delta}$ films in the spectral region from 100 μm to 1 μm with response times depending on film thickness and explained by heat transfer to the substrate [10]. All these observations are consistent with our results for thallium compound films [13] and indicate a critical frequency for $YBa_2Cu_3O_{7-\delta}$ films at photon energies $\nu_{crit} \leq 3$ THz. In several works, fast response at high photon energies in relatively thin films has been attributed to nonthermal processes [11, 12]. However, all response times observed at high photon energies are consistent with a cooling time $\tau \simeq d^2/2k$. Heat escape, which occurs within several ns for a thin film of 100 nm thickness, lasts a time of ~ 1 μs for a film of 1 μm thickness. For a clear distinction between «slow» bolometric signal generating processes and processes leading to fast response use of thick films ($d \sim 1$ μm) is favourable.

The present result $2\Delta(0)/k_B T_c \simeq 2$ can be compared to data obtained from FIR reflectivity measurements. Studies of the (a, b)-plane conductivity of $YBa_2Cu_3O_{7-\delta}$ suggest an energy gap $2\Delta(0)/k_B T_c \simeq 6$ to 8 [2, 17], and a second smaller energy gap with $2\Delta(0)/k_B T_c \simeq 2$ to 4 [2, 18]. From a measurement with c-axis polarization $2\Delta(0)/k_B T_c \simeq 3$ was reported [3]. In measurements on ceramic $Tl_2Ba_2CaCu_2O_8$ [19] and on epitaxial $Tl_2Ba_2CaCu_2O_8$ films [20], it has been found difficult to assign a feature in the reflectivity spectrum to an energy gap. As the onset of slow optical response probes the smallest energy gap in granular materials, the small value of Δ may be related to Cooper pair breaking for carriers propagating in directions oblique to the (a, b)-plane as well as to a reduced gap in the surface layer of grains. It has been shown that Δ substantially decreases on the surface of high-T_c superconductors even for $T \ll T_c$ due to the short coherence length [21].

In summary we reported on fast far-infrared optical response for current-carrying granular $Tl_2Ba_2CaCu_2O_8$ films at low temperature. We attribute the fast optical response to currents induced by the infrared field in weak links at grain boundaries. The fast signal occurs below a critical frequency that may be due to a superconducting energy gap, possibly along the c-direction. The critical frequency corresponds to an energy gap of $2\Delta(0)/k_B T_c \simeq 2$. Our results also demonstrate that granular thin films containing weak links are well suitable as fast FIR detectors with most likely subnanosecond response times.

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