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PERSISTENT PHOTOCONDUCTIVITY IN INSULATING $YBa_2Cu_3O_X$ THIN FILMS.

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Abstract

The illumination of $YBa_2Cu_3O_x$ thin films increases the critical temperature and decreases the normal state resistivities *if and only if* the films are oxygen deficient. For films with an oxygen content x > 6.4 the persistent photoconductivity (PPC) is small, but clearly present.

Oxygen deficient a-axis oriented $EuBa_2Cu_3O_x$ thin films exhibit the PPC effect as well, and the magnitude of the effect seems to be comparable to what is observed in c-axis films with the same oxygen content.

For YBCO films with x < 6.4, the photoconductivity is substantially enhanced when the oxygen content is lowered and increases exponentially when $x \to 6.0$.

1. Introduction

Recently, persistent photoconductivity (PPC) [1] and photoinduced superconductivity (PSC) [2] were discovered in the high T_c superconductors $RBa_2Cu_3O_x$ (with R = Y, Eu, Gd). When an oxygen depleted (x < 7) c-axis oriented thin film is illuminated with visible light, its electrical resistance drops and its critical temperature increases. At the same time the c-axis contracts [3], and the Hall coefficient decreases during illumination, which indicates an increase in the number of charge carriers [4]. These effects are persistent if the film is kept at low temperatures. At room temperature, the properties of the film relax to their original state over a period of several days [4].

In earlier work, we showed that an insulating film with an oxygen content near the metal-insulator transition can be excited to become superconducting [2], and that the magnitude of the PPC effect increases as the sample approaches the metal-insulator transition [2,4-6]. Moreover, oxygen vacancies in the chain layers seem to be essential since no persistent photoconductivity could be measured in fully oxidized $RBa_2Cu_3O_x$ (R = Y or Gd), nor in $Bi_2Sr_2CaCu_2O_{8+\delta}$ films with different oxygen contents or $La_{2-x}Sr_xCuO_4$ films with different Sr doping levels [6].

In this paper, we report on the photoinduced effect in an a-axis oriented $EuBa_2Cu_3O_x$ thin film and on the magnitude of the photoinduced phenomena in $YBa_2Cu_3O_x$ thin films with $x < 6.4$ after continuous-wave laser illuminations at 100 K. We show that, in contrast to the results obtained by Kudinov et al. [5], the PPC effect increases as $x \rightarrow 6.0$. The crystallographic orientation of the film seems to have no influence on the magnitude of the effect.

2. Experimental

Fully oxidized c-axis oriented $YBa_2Cu_3O_7$ thin films were deposited on MgO(100) substrates using a planar magnetron sputtering system in the 90° off-axis sputtering configuration at a deposition temperature of 740 °C [7]. These as-prepared films have critical temperatures of 87 - 91 K and superconducting transition widths < 2 K.

The a-axis oriented $EuBa_2Cu_3O_x$ film was deposited on a $SrTiO_3(100)$ substrate by dc sputtering from a single stoichiometric target. In order to obtain an a-axis orientation of the film, a two-step substrate heating procedure was used [8]. An a-axis oriented seed layer was grown at ± 660 °C during the first minutes, afterwards the substrate temperature was raised to 720 °C [9]. By this method a-axis oriented films with high critical temperatures ($T_c > 80$ K) can be obtained [10].

Oxygen deficient films are prepared using a method based on published oxygen partial pressure - temperature phase diagrams for bulk material [11]. For the YBCO films (c-axis), the phase diagram published by Tetenbaum et al. [12] was used, which for oxygen contents $x > 6.4$ results in critical temperatures that coincide with those obtained in bulk material [13]. The values of x for $x < 6.4$ are only nominal ones. Due to the lack of published phase diagrams for EuBCO, this film was prepared following an earlier published phase diagram for YBCO [14]. This phase diagram, however, results in YBCO films and bulk material with T_c's which are too low with respect to their nominal x-value (shift of ≈ 0.1 in x), which may also be the case for the EuBCO films. In order to prevent adverse reactions with contaminants or film decomposition, we kept the temperature below 600 °C. After each oxygen depletion, the sample was kept at room temperature for at least three days in order to allow a complete relaxation [15].

The YBCO film was patterned into a four point pattern (dimensions of the bridge: 40 μm x 1 mm) using conventional photolithography and wet etching. Wire bonding was used to connect the electrical leads to the sample. These contacts can give rather high contact resistances, but have the advantage that the contacts can be easily removed before the different heat treatments that are performed on the same film. The EuBCO film was unpatterned and therefore a four lead Montgomery technique [16] was used to obtain the resistivity values.

The illuminations were performed in a He flow cryostat with optical windows at 100 K using an Ar-ion laser (λ = 514 nm) for the YBCO films, and a halogen lamp for the EuBCO film. For the YBCO films, the measured power density on the sample surface was 1.83 W/cm^2, corresponding to a total photon density Q ≅ 1.4 x 10^{23} /cm^2 after 8 hours of illumination. During the illumination the sample temperature increased by at maximum 5-6 K. The resistance versus temperature curves were measured in-situ before and after illumination during the warming-up cycle and up to temperatures not exceeding 250 K.

3. Results

Fig. 1 shows the resistivity as a function of temperature for a single YBCO film with different oxygen contents $x \leq 6.4$. The critical temperature of the as-prepared film was 87 K and the resistivity at 300 K was 0.2 mΩcm. For x=6.4, the film has a superconducting transition at 27 K. When the oxygen content is reduced to x = 6.3, the sample is metallic at high temperatures and semiconducting at low temperatures. For oxygen contents lower then 6.2, the samples are insulating in the whole temperature region (5 - 300 K). For the low oxygen contents the resistance could be measured only up to values comparable to the input-resistance of the voltmeter.

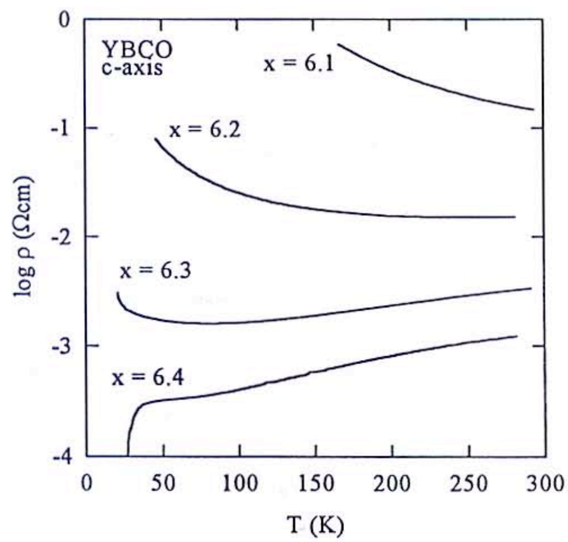

Figure 1. The resistivity versus temperature for a c-axis oriented $YBa_2Cu_3O_x$ film with different oxygen contents.

Fig. 2 shows the resistivity as a function of temperature before and after illumination for the YBCO film with x=6.4 (Fig. 2a) and x=6.2 (Fig. 2b). After the illumination, the critical temperature of the x = 6.4 film increases by 6 K, and its resistivity drops over the entire temperature range (from 0.40 mΩcm to 0.32 mΩcm at 100 K). In the insulating x = 6.2 sample, the resistivity decreases by one order of magnitude over the entire temperature range. After several days of annealing at room temperature, the original resistivity values are recovered.

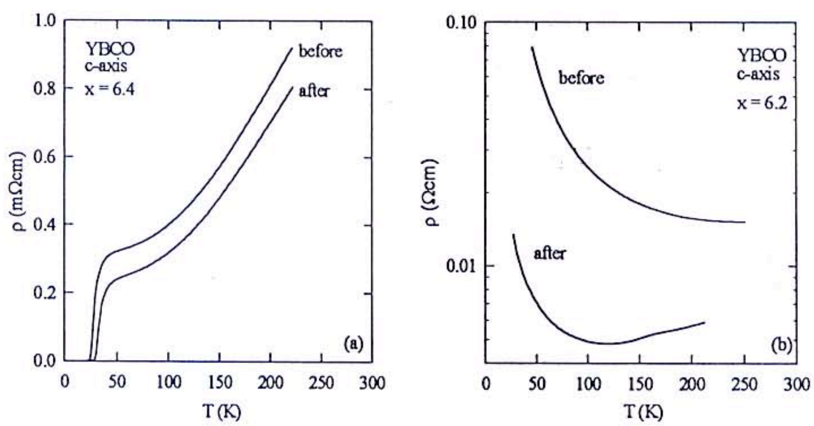

Figure 2. The resistivity versus temperature for the x = 6.4 YBCO film (a) and the x = 6.2 YBCO film (b) before and after illumination.

The resistivity before and after illumination for the a-axis oriented EuBCO film with nominal x = 6.5 is shown in Fig. 3. The resistivity before illumination shows a clear superconducting onset at low temperatures, but is not fully superconducting above 8 K. After illumination the sample is fully superconducting at 10 K. The resistivity of the film also decrease due to the illumination (33 % at 100 K). This drop is comparable to the one seen in c-axis films with the same nominal oxygen content, prepared in exactly the same way.

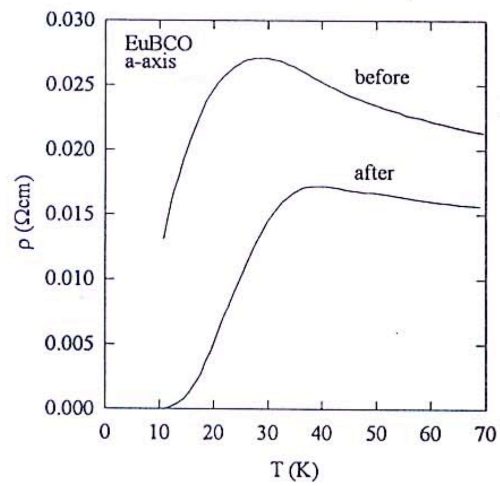

Figure 3. Resistivity as a function of temperature for an a-axis oriented $EuBa_2Cu_3O_x$ film before and after illumination.

The absolute increase of the in-plane conductivity at 100 K for the c-axis YBCO film, $\delta\sigma = \sigma_f - \sigma_i$, is shown in Fig. 4. We define σ_i and σ_f as the conductivity before and after illumination. Also shown is the conductivity before illumination σ_i. We included in Fig. 4 the results published elsewhere which were obtained on a second film with $x > 6.35$ [6]. A clear maximum can be seen around $x = 6.4$, which is above the metal-insulator transition for the films. However, at lower oxygen concentrations, σ_i is negligibly small compared to σ_f and therefore, their difference $\delta\sigma$ is dominated by σ_f. Under the assumption that σ_f is proportional to σ_i, the low temperature behaviour reflects the characteristic decrease in conductivity as the samples become insulating.

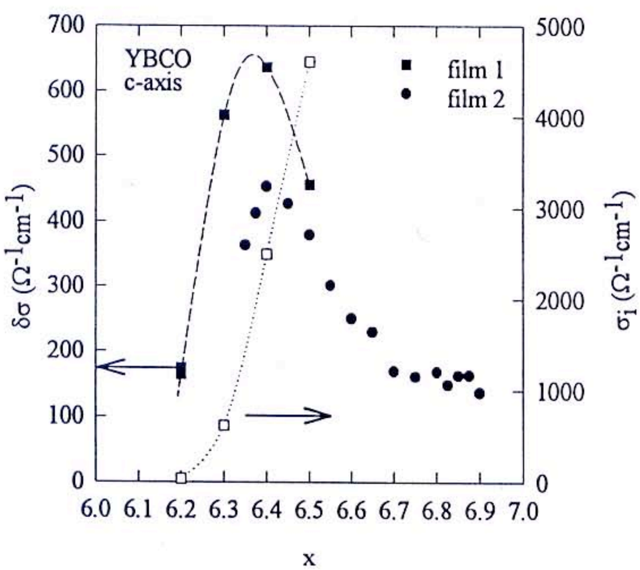

Figure 4. Absolute increase in conductivity versus oxygen concentration of two c-axis oriented $YBa_2Cu_3O_x$ films at 100 K. Also shown are the dark conductivity σ_i values for $x \leq 6.4$.

The magnitude of the PPC effect in insulating films is best characterised by the increase in conductivity by illumination at a fixed temperature, normalized by the conductivity before illumination ($\delta\sigma/\sigma_i$). This normalization removes the inherent decrease in conductivity as the oxygen content is decreased. The results for measurements at two temperatures (100 K and 210 K) in the two films are shown in Figure 5. It is clear that the PPC effect is greatly enhanced at low oxygen concentrations and seems to increase faster than exponentially for $x \rightarrow 6.0$. The insert shows the results for $x > 6.35$, clearly illustrating that the effect is present as soon as $x < 7$.

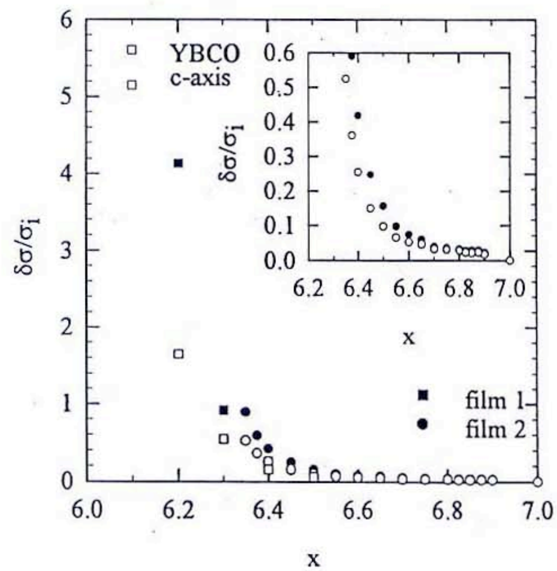

Figure 5. Absolute increase in conductivity versus oxygen concentration measured at two different temperatures (T = 100 K: solid symbols, and T = 210 K: open symbols). The squares are data obtained in film 1 for $x \leq 6.4$, and the circles are data from film 2 for $x \geq 6.35$. The inset shows details of the absolute increase in conductivity versus oxygen content for the higher x values.

In order to check if the quality of our film did not degrade by the oxygen removals, we reoxidized the samples between the different oxygen depletion treatments. Even after several treatments, the original $x = 7$ curve was recovered. Therefore, any change of σ and T_c is due to a reversible change in the oxygen concentration and not to adverse chemical reactions during processing.

Between $x = 7$ and $x = 6.4$ our experimental results are similar to those reported by Kudinov et al. [5]. However, for $x < 6.4$ Kudinov observes a continuous decrease of the PPC effect (Ref. [5], Fig. 11), in complete disagreement with our present results. The origin of this difference could arise from the way the experiment was performed. Kudinov et al. [5] illuminated at 300 K, while our data were obtained at 100 K. However, a room temperature illumination of our $x = 6.1$ sample resulted in a 50 % decrease of the resistivity, Kudinov et al. claim there is no change [5]. Kudinov et al. quenched their samples from high temperature during oxygen removal treatment instead of maintaining thermodynamic equilibrium down to low temperatures. This quenching procedure might suppress the PPC effect in some unknown way. Finally, Kudinov et al. did not reoxygenate their samples to check for adverse chemical reactions. We therefore conclude that either their samples may have been contaminated during oxygen treatment or the effect is suppressed in quenched samples in an unknown fashion.

4. Conclusions

Persistent photoconductivity occurs in $RBa_2Cu_3O_x$ films as soon as the material is oxygen deficient ($x < 7$). In contrast to what was earlier published by Kudinov et al. [5], we find that the PPC effect is greatly enhanced for $x \rightarrow 6.0$ in $YBa_2Cu_3O_x$ films. Furthermore, the orientation of the film seems to have no influence on the magnitude of the effect.

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