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Photoexcitation effects in $YBa_2Cu_3O_x$

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Abstract

We show through photoexcitation experiments carried out on $YBa_2Cu_3O_x$ c-axis oriented films that photoinduced phenomena are not restricted to insulating samples. Superconducting films with critical temperatures varying between 2 K and 40 K show a clear enhancement of the superconducting transition after illumination. The increase in T_c is accompanied by a corresponding decrease in the resistivity ρ, and an increase in the Hall coefficient R_H, indicative of an increased carrier density. Furthermore, the carrier mobility is also affected by illumination.

1. Introduction

Photoconductivity phenomena have a long history and occur in a variety of insulating and semiconducting materials. The existence of a metal-insulator transition (MIT) in high T_c superconductors opened up the possibility to perform photoexcitation experiments in these materials. In this framework, transient photoinduced changes of more than ten orders of magnitude in the surface resistivity of $YBa_2Cu_3O_x$ (YBCO) single crystals have been reported [1]. Persistent photoconductivity measurements in insulating YBCO films [2] clearly showed that laser illumination induces a systematic decrease with long relaxation times in the electrical resistivity $\rho(T)$ of insulating YBCO films. Other photoinduced phenomena have been observed by Raman scattering measurements [3, 4].

In this paper we will review the experimental results obtained by illuminating oxygen deficient YBCO films [5]. It will be shown that i) the decrease in resistivity in superconducting films is accompanied by a simultaneous increase in T_c; ii) the carrier density as well as the carrier mobility changes; and iii) after illumination a long relaxation time at room temperature is observed. It is interesting to note that these photoinduced phenomena have been observed - to our knowledge for the first time - in *metallic* films with critical temperatures as high as 40 K, although the relative changes are larger for films with lower oxygen contents. This experimental observation seems to raise doubts about the proposed mechanism based on the existence of a gap in the bandstructure [2], analogous to photoconductivity effects in semiconductors.

2. Experimental

High quality superconducting YBCO films are prepared using 90° off- axis magnetron sputtering from a single stoichiometric target. Details of the film preparation technique can be found elsewhere [6]. In order to reduce the oxygen content of the films, a special heat treatment in a controlled oxygen pressure is applied, based on the oxygen pressure - temperature (P_{O_2} - T) phase diagram. This procedure consists of heating up the film - which is placed inside a small YBCO ceramic box - at a constant oxygen pressure $P_{O_2} = 10$ Torr to the temperature T in the phase diagram which corresponds to the desired oxygen content x_n. After a few hours of stabilisation, the film is slowly cooled while the oxygen pressure is decreased in order to follow the x_n phase line. When P_{O_2} is lower than 75 mTorr, the film is quenched down to room temperature. This method has been shown to be a reliable, reproducible and reversible technique for controlling the oxygen content in thin YBCO films [7].

Photoconductivity and photoinduced Hall effect measurements were performed on patterned 1000 Å thick oxygen deficient YBCO films. The resistivity was measured using the standard four probe technique, with a current density of 20 A/cm^2. The room temperature Hall constant R_H was measured with a current density of 2000 A/cm^2 in a 6 kGauss magnetic field, using the standard field inversion technique. Care was taken to place the Au voltage contact pads outside the current flow in order to avoid any possible spurious photoconducting contribution from the metal-YBCO contact.

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Two types of light sources were used for the film illumination : an Ar ion laser with a series of lines in the range 454.5 nm $< \lambda <$ 514.5 nm and a total output power of 6 W, and an ordinary halogen white light source. During laser light illumination the samples were immersed in liquid nitrogen, while for the halogen lamp illumination, they were kept at room temperature. The absence of sudden changes in the resistivity immediately after turning off the light source indicates that in both cases (laser or halogen lamp illumination) the heating caused by illumination is negligible.

3. Results and discussion

Figure 1 shows the resistive transition before and immediately after laser light illumination for four YBCO films with different nominal oxygen contents. For all the films illumination induces a decrease in resistivity and - except for the insulating film - an increase in T_c. The decrease in resistivity of the superconducting films, monitored in situ during illumination at 77 K, is shown explicitly in the inset of Fig. 1 (a). Notice that the photoinduced changes in ρ are most pronounced for the films with a lower initial T_c. The insets in Figs. 1 (b), (c) and (d) show the region near T_c in expanded scale. Again the relative change in T_c is larger for the films with a lower initial T_c.

Photoconductivity measurements have also been performed using an ordinary halogen lamp. The changes in the resistivity, monitored in situ during and after illumination at room temperature, are presented in Fig. 2 (a) for an $x_n = 6.5$ YBCO film. A pronounced photoinduced change in ρ is indeed observed. Notice the difference in time scale between the excitation process and its relaxation. The long relaxation times will be discussed in more detail further in the paper.

In order to see if the observed photoconductivity effect can be directly related to an increase in carrier density, we measured the Hall coefficient R_H during and after the halogen lamp illumination. The evolution of R_H for the $x_n = 6.5$ YBCO film is shown in Fig. 2 (b). It is clear that ρ and R_H have the same time dependence during and after illumination. Assuming that the Hall coefficient is inversely proportional to the carrier density, the results shown in Fig. 2 (b) indicate that photocarriers are generated during illumination. Moreover, by computing the carrier mobility $\mu = R_H/\rho$ (shown in Fig. 2 (c)), it is clear that the observed changes in resistivity are not only due to changes in carrier density, but that the mobility is also affected by illumination and contributes to variations in ρ.

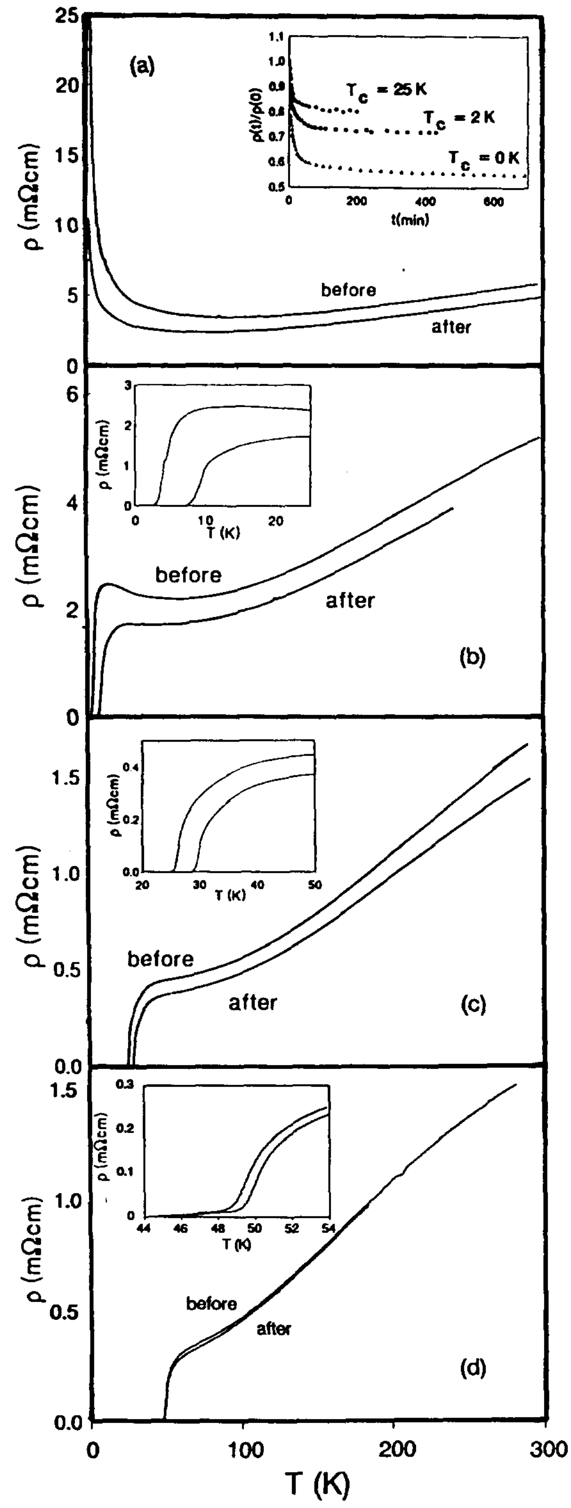

Figure 1 : Electrical resistivity vs temperature before and immediately after laser illumination for YBCO films with nominal oxygen stoichiometries (a) $x_n = 6.5$, (b) $x_n = 6.55$, (c) $x_n = 6.6$ and (d) $x_n = 6.6$. The inset in (a) shows the time evolution of the resistivity at 77 K during illumination for samples a, b and c. The insets in (b), (c) and (d) show the region near T_c in expanded scale.

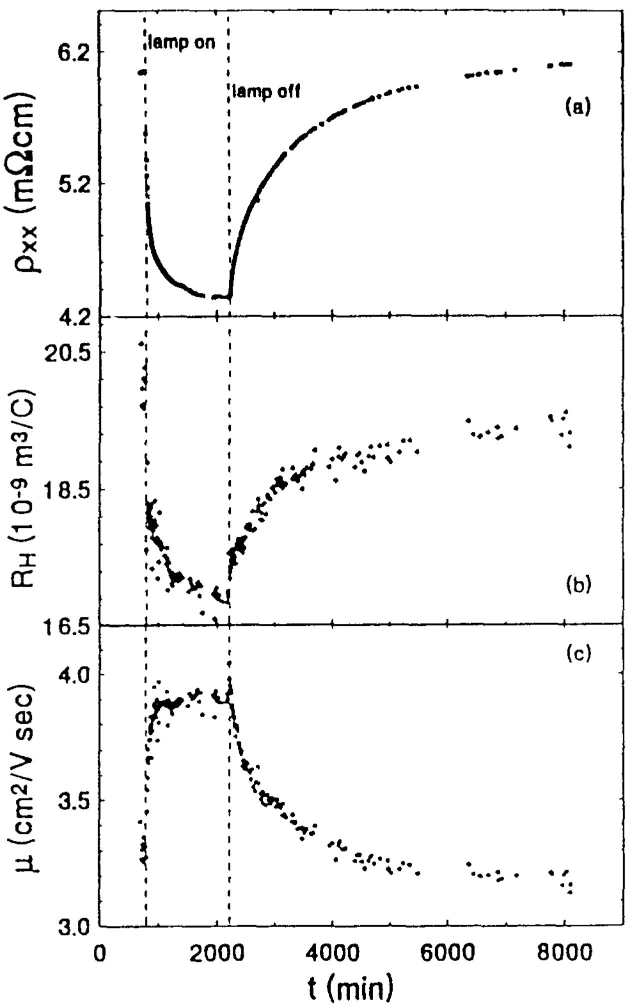

Figure 2 : Time dependence at room temperature during and after halogen white light illumination of (a) the electrical resistivity ρ, (b) Hall coefficient R_H, and (c) mobility μ, for an $x_n = 6.5$ YBCO film.

Similar mobility variations were also obtained from ρ and R_H measurements during relaxation experiments at room temperature in films illuminated by laser light at liquid N_2 temperature. Figure 3 shows the mobility versus time for films with different oxygen contents. The data were normalised with respect to their initial values (which were independent within 10 % upon x_n). It is clear from these data that the relative photoinduced changes in the mobility are also larger for smaller x_n values.

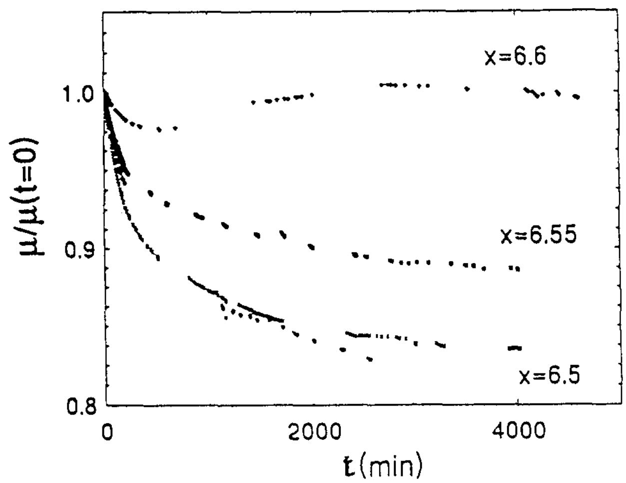

Figure 3 : Time dependence at room temperature after Ar ion laser light illumination at 77 K of the mobility μ for YBCO films of different oxygen content x_n. Values have been normalised to the initial value, which for all samples is 4.2 ± 0.3 cm^2/V.sec.

The microscopic mechanism underlying the observed photoinduced phenomena is not clear at present. Two different ideas seem to emerge. The first assumes that electron-hole pairs in the CuO_2 plane are photogenerated. The electrons move to the basal CuO_x plane, where they are captured by the oxygen vacancies. The holes remain free and cause an increase in the nonequilibrium carrier density [2].

In the second picture, photoassisted oxygen ordering takes place. The problem of oxygen ordering in oxygen deficient YBCO samples has attracted theoretical [8] and experimental [9] attention, and is believed to induce a transfer of holes to the CuO_2 planes [10].

In order to study the possible relation between photoinduced phenomena and oxygen ordering, resistivity and Hall effect measurements were carried out as a function of annealing time t_a on quenched oxygen deficient YBCO films. During these aging experiments, the sample remained isolated from any light source. The aging of ρ and R_H for an $x_n = 6.6$ film is shown in Fig. 4. Similar to the illumination process, the aging induces a decrease of the resistivity and of the Hall coefficient. The mobility (inset of Fig. 4) however decreases during aging, while it increases during illumination. The decrease in ρ (t_a) and R_H (t_a) seen in this non-illuminated film has been interpreted as a consequence of oxygen ordering effects [11].

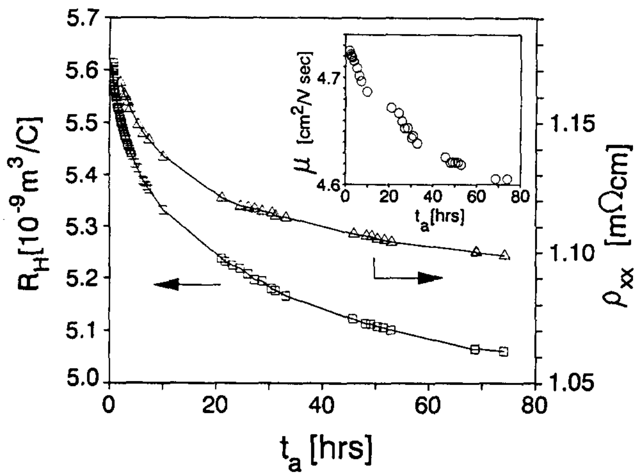

Figure 4 : Time dependence at room temperature, starting immediately after deoxygenation, of the electrical resistivity ρ, and Hall coefficient R_H, for a non-illuminated x_n = 6.6 YBCO film. Inset shows the time dependence of the mobility μ. Solid lines are guides to the eye.

In view of the result that oxygen ordering is induced during aging at room temperature, the observation of photoinduced phenomena even during room temperature illumination raises doubts about photoassisted oxygen ordering as the operating mechanism. Nevertheless, the striking similarity between the room temperature relaxation times after illumination (Figs. 2 and 3) and after quenching seems to indicate that - irrespective of the photoconductivity mechanism - the decay of the photogenerated carriers is governed by a thermal diffusion process of oxygen atoms. In order to understand the microscopic excitation mechanism, more work is clearly necessary. Finally, in order to differentiate between possible microscopic mechanisms it is necessary to determine exactly where the light is absorbed.

4. Conclusions

In summary, we observed substantial photoinduced changes in the resistivity, the critical temperature and the Hall coefficient of metallic oxygen deficient YBCO films. The amplitude of these changes increases with decreasing oxygen content. Relaxation experiments at room temperature show that the relaxation times of the resistivity and the Hall coefficient are comparable to the ones measured in quenched non-illuminated oxygen deficient films.

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