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Photoinduced Enhancement of Superconductivity

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The photoinduced enhancement of superconductivity in $RBa_2Cu_3O_x$ (R = rare earth or yttrium) and $Pr_yR_{1-y}Ba_2Cu_3O_x$ was explored through temperature-dependent resistivity, Hall coefficient and mobility, and x-ray diffraction measurements. The increases in T_c are enhanced near the metal-insulator transition, although photoinduced changes always exist in oxygen-deficient samples. Several explanations, including intergrain Josephson coupling, photoassisted oxygen ordering, and the trapping of photogenerated electrons in oxygen vacancies, are discussed.

KEY WORDS: Superconductivity; thin films; photoconduction.

The interest in photoinduced superconductivity in $RBa_2Cu_3O_x$ (R = rare earth or Y) was sparked by the discovery of transient photoconductivity in oxygen-deficient YBCO single crystals by Yu *et al.* in 1989. In their experiments, Yu *et al.* observed an increase of several orders of magnitude in the crystal's transient photoconductivity at 50 K when excited by a picosecond pulsed laser [1]. Shortly thereafter, V. Kudinov and co-workers observed a *persistent* photoinduced decrease in the resistivity of oxygen-deficient YBCO thin films at temperatures below 270 K. Some of these films seemed to display photoinduced superconductivity, that is, the resistivity characteristics changed from semiconducting- to superconducting-like by illumination with an Ar laser [2]. For the superconducting samples, the transition widths were large enough that it was unclear whether the observed effects were a result of simple intergrain Josephson coupling, or genuine intrinsic photoexcited superconductivity. Such intergrain coupling effects have been observed in In-CdS granular films by Deutscher and Rappaport in 1979, where the transition widths of the films were significantly decreased by photoexcitation [3].

Motivated by Kudinov's results, we recently demonstrated that oxygen-deficient semiconducting YBCO films indeed show a sizable decrease in the normal-state resistivity in the temperature range between 4 and 300 K (see Fig. 1a). More importantly, in *metallic*, superconducting samples, the T_c was significantly enhanced by as much as $\sim$5 K, with the transition width remaining unchanged and the normal-state resistivity parallel to the *ab* plane, ρ_{ab}, decreasing throughout the whole temperature range (Fig. 1b–c) [4]. These effects were found to be persistent below 270 K, and could be induced by either an Ar laser or a halogen lamp. At room temperature, the photoinduced effects decay over a period of days, and moreover, are accompanied by a large decrease in the Hall coefficient R_H (Fig. 2) [5]. The mobility, defined as $\mu \equiv R_H/\rho_{ab}$, was also found to change with illumination (Fig. 2c), which indicates that the effect cannot be explained by only an increase in the number of carriers deduced from a simple one-band model. Further studies revealed that the effect becomes systematically weaker with increasing oxygen concentrations [6].

These studies indicate that the photoinduced transport effects are similar to those observed in oxygen-deficient RBCO films when their oxygen

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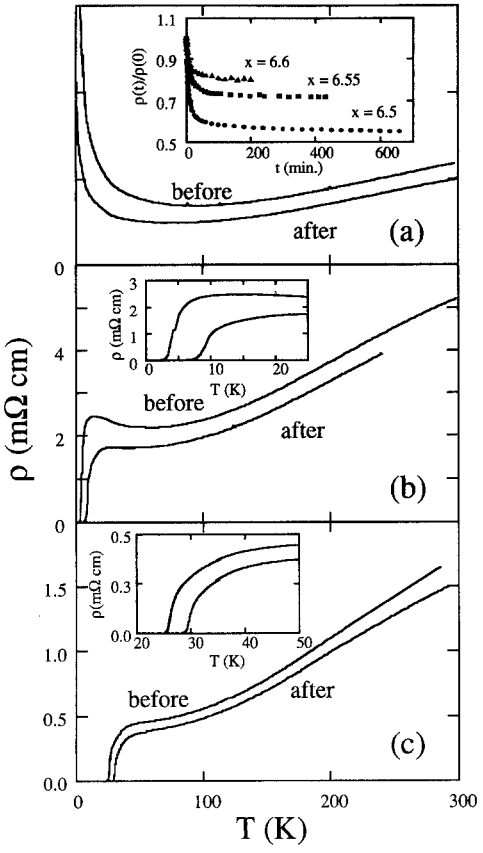

Fig. 1. Electrical resistivities ρ_{ab} as a function of temperature T, before and immediately after laser illumination of YBCO films with oxygen concentrations $x = 6.5$ (a), 6.55 (b), and 6.6 (c). The inset in (a) shows the normalized electrical resistivity, $\rho(t)/\rho(0)$, at 77 K as a function of time, t, for these samples. The insets in (b) and (c) show the region near T_c in an expanded scale [4].

content is increased [7], when they are quenched from high temperatures to room temperature [8], and when the Pr concentration in Pr/RBCO is decreased [9]. Hence, this raised a number of experimental questions, such as the dependence of the photoinduced effects on the metal–insulator (M–I) transition, oxygen deficiency, and the possible structural changes.

The connection between photoexcitation and the M–I transition was recently explored using $Pr_yGd_{1-y}Ba_2Cu_3O_x$ phase-spread alloy films simultaneously prepared on the same substrate [10]. Figure 3 shows the effect of illumination on the resistivities of two fully oxygenated films with different Pr concentrations [11]. No significant changes in the resistivities or the T_c's of these films are detected. This result may be compared with that of Fig. 1a, where a semiconducting, oxygen-deficient RBCO films shows a

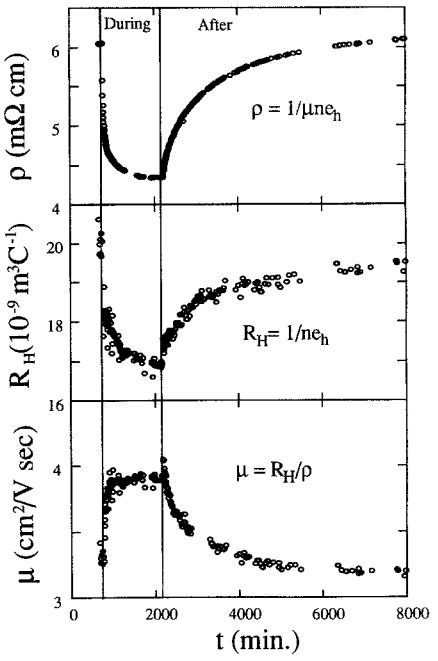

Fig. 2. Time dependence at room temperature after halogen white light illumination of the electrical resistivity ρ_{ab}, Hall coefficient R_H, and Hall mobility $\mu = R_H/\rho_{ab}$, for a semiconducting $YBa_2Cu_3O_{6.5}$ film [5].

~40% decrease in its resistivity for the whole temperature range. Conversely, in Fig. 4 we show the effects of illumination on the resistivities of four oxygen-deficient Pr/GdBCO films with $0.05 < y < 0.23$ and

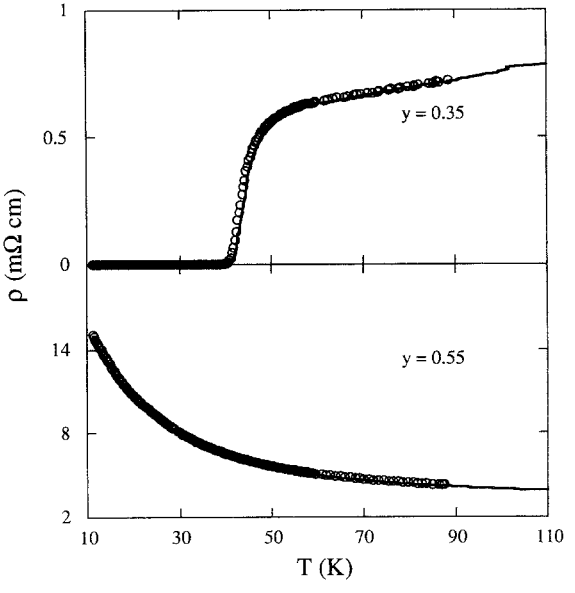

Fig. 3. Resistivities of two fully oxygenated $Pr_yGd_{1-y}Ba_2Cu_3O_{7.0}$ simultaneously grown films on the same substrate before (○) and after illumination (solid lines) at 95 K for ~12 h. [11].

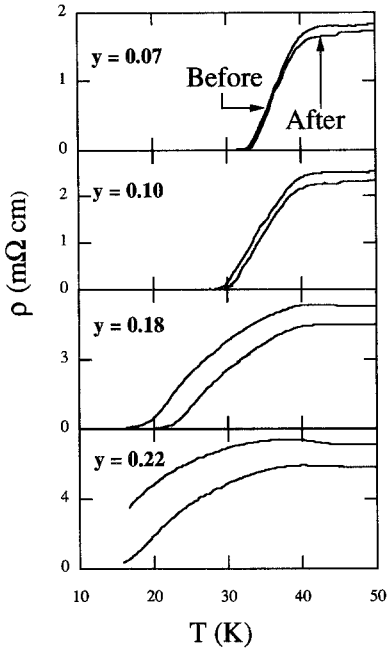

Fig. 4. Resistivities of a set of oxygen-deficient $Pr_yGd_{1-y}Ba_2Cu_3O_x$ films (with $x \sim 6.7$) before and after illumination [11].

$x \approx 6.7$. A clear enhancement of superconductivity, characterized by an increase of the T_c and a decrease of the normal-state resistivities, can be observed in all the films. Although the increases in T_c are enhanced near the M–I transition, the proximity to the M–I transition is not a sufficient condition for the observation of photoinduced enhancement of superconductivity in the absence of oxygen vacancies. It should be kept in mind, however, that this enhancement in the spread alloy films could be simply explained if larger Pr concentrations induce larger oxygen deficiencies. In any case, it is the presence of oxygen vacancies which is clearly the crucial aspect of the effect.

Photoinduced structural changes, deduced from the [00 10] x-ray diffraction peak of c-axis oriented films, were studied in the $x = 6.5$, YBCO sample shown in Fig. 1a. In Fig. 5 the fractional changes in the c-axis lattice parameter ($\Delta c/c$) and resistivity ($\Delta\rho/\rho$) as functions of time are shown during and after illumination. Notice that $\Delta c/c < 0$, which is the opposite of what would be expected from simple thermal expansion effects. Furthermore, the resistivity and lattice parameter changes appear to be correlated. Further structural information can be obtained by refining the full Θ–2Θ scans of this sample with a method developed by Fullerton and co-workers [12] before and immediately after illumination, as shown

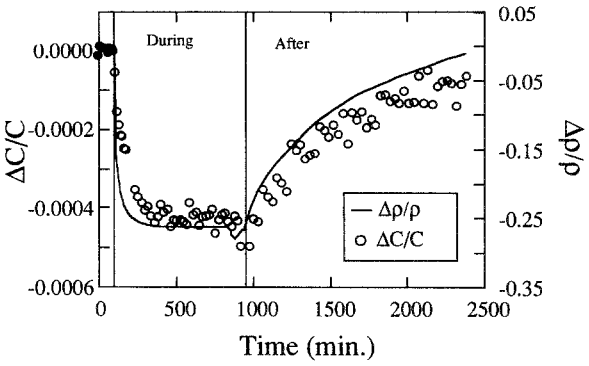

Fig. 5. Fractional changes $\Delta c/c$ of the c-axis lattice parameter (O) and resistivity $\Delta\rho/\rho$ (solid line) vs. time in a semiconducting 1000 Å thick $YBa_2Cu_3O_{6.5}$ film before, during, and after illumination [11].

in Fig. 6a and b. Since the two scans are almost identical, in Fig. 6c we have plotted their subtraction, where the c-axis contraction is evident. As mentioned in [10] the results of the fits indicate that most of the contraction occurs in the $BaO–CuO_x$ (Ba–Cu1) interplanar distance parallel to the c-axis. We note that as oxygen is added to oxygen-deficient YBCO single crystals, neutron diffraction studies show that the c-axis contracts. This contraction occurs precisely in the Ba-Cu1 interplanar distance, while the other two interplanar

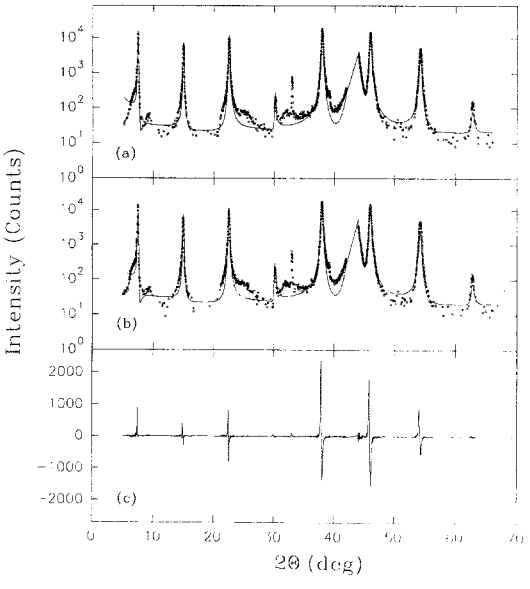

Fig. 6. (a, b) Data (O) and simulation (solid line) of scans performed before and 9 min after 12 h of illumination on a semiconducting 1000 Å thick $YBa_2Cu_3O_{6.5}$ film. The peak near $2\Theta = 33°$ corresponds to In used for the resistivity contacts. The large peak at $2\Theta = 42.9°$ (which is cut off) corresponds to the [002] MgO substrate. (c) Subtraction of the data in (b) from (a) [11].

distances, R–Cu2 and Cu2–Ba, expand [13,14]. Similar contractions of the *c*-axis in quenched oxygen-deficient films have also been observed [8,15].

Three different explanations of the photoinduced enhancement of superconductivity have emerged. In the first picture, the effect is explained by assuming that the film is composed of superconducting grains surrounded by nonsuperconducting, photosensitive materials, as in the In–CdS system studied in [3]. Photosensitive Josephson coupling between superconducting grains would then be the responsible mechanism of the photoinduced enhancement of superconductivity, and would be characterized by a significant decrease in the transition width. This explanation is not attractive, however, since in our films the resistive transition moves to higher temperatures in a parallel fashion, and its width does not appear to change much. A second explanation relies on photoinduced oxygen ordering, which would cause an increase in the number of carriers in the CuO_2 planes [16]. This mechanism is attractive because changes in transport and structural properties similar to those produced by photoexcitation have been observed in oxygen-deficient films and single crystals quenched from high temperatures (~400°C) to room temperature [8,15]. The third explanation relies on the photoinduced creation of electron–hole pairs, with the oxygen vacancies acting as deep electron traps [17]. This would leave a free hole that would increase the carrier density. The structural changes would then be due to the local distortions caused by the trapping of electrons. A similar effect has been observed in $Al_xGa_{1-x}As$ with impurities (or DX centers) acting as deep electron traps. Due to the long lifetime of the effect at room temperature, the binding energies involved in the effects in YBCO would have to be at least a factor of 5 larger than those observed in GaAs. Hence, the trap energy must be at least ~1 eV. Because superconductivity may be enhanced in metallic samples, the latter explanation is plausible if light causes interband transitions. However, at the present time there is no direct experimental proof for this mechanism.

In summary, the illumination of oxygen-deficient RBCO films causes structural and transport changes similar to those observed when the oxygen concentration in oxygen-deficient RBCO is increased. This effect can only be observed if oxygen vacancies are present in the material and is enhanced by the proximity to the M–I transition. The microscopic mechanism responsible for the photoinduced enhancement of superconductivity so far has not been elucidated.

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