Photoinduced enhancement of superconductivity

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We show clear and conclusive experimental evidence for the enhancement of superconductivity in $GdBa_2Cu_3O_x$ and $YBa_2Cu_3O_x$ thin films by photoexcitation. Upon laser illumination the critical temperature increases while the resistivity of the material decreases. The relaxation back to the original state is very slow, of the order of days at room temperature. The existence of this effect opens the possibility of fabricating weak-link devices with *in situ* tunable superconducting characteristics.

The reliable preparation of weak links is the key obstacle for the development of Josephson-effect-based high-temperature superconducting electronics. Since the coherence length in high T_c superconductors is very short, of the order of nanometers, photolithography at this length scale is beyond the capabilities of present day technology. It is therefore desirable to develop methods for *in situ* change of the superconducting properties of thin films.

The interaction of superconductivity with lasers has been studied extensively in conventional super- conductors.[1,2] The effect for these materials consists in a decrease of all superconducting properties. It has been shown recently that the photoexcitation of $YBa_2Cu_3O_x$ (YBCO) reduces the resistivity of the material, that the effect persists for long times of the order of days, and that "prolonged irradiation leaded to a *seemingly* complete loss of resistivity below 5 K."[3] Recent transient photoinduced conductivity experiments[4] have also claimed a decrease in the resistivity of insulating YBCO single crystals although in a much shorter time scale, of the order of nanoseconds. Raman scattering measurements in the fully oxygenated photoexcited sample, indicate the appearance of long-lived nominally forbidden phonon modes.[5] On the other hand, photoinduced electronic absorption was observed in insulating YBCO by infrared absorption.[6,7]

The present experiments conclusively show that photoexcitation induces and enhances superconductivity in $RBa_2Cu_3O_x$ (R = Gd, Y) thin films of well-defined and controlled oxygen stoichiometry. Sharp and well-defined superconducting transitions are shifted up in temperature in a clear fashion. The zero-resistance state is induced at higher temperatures by the laser illumination.

High quality YBCO and GdBCO epitaxial films were prepared by two different *in situ* sputtering methods[8,9] on $SrTiO_3$ and MgO. The details of the not aligned chopped power oscillatory (NACHOS)[8] and standard 90° off-axis[9] preparation methods are described elsewhere. Briefly, the films were prepared at ~750 °C in a mixture of argon and oxygen, and then cooled to room temperature (RT) through an oxygen doping sequence.

Figure 1 shows the resistivity as a function of temperature for a 200-nm-thick GdBCO insulating film, before and after laser illumination. The same behavior reported earlier by Kudinov *et al.*[3] is observed; i.e., a drop of the resistivity in all the temperature range and the appearance of an abrupt decrease in the resistivity at low temperatures which may eventually culminate in a zero-resistance state below 1.5 K. This result and those of Ref. 3, which indicate the possibility of a photoinduced enhancement of superconductivity, encouraged us to investigate this effect under more controlled circumstances. We conducted experiments on insulating GdBCO films and YBCO films with different oxygen contents on both sides of the metal-insulator transition.

The YBCO oxygen deficient samples were prepared by thermal treatment in a controlled oxygen environment.[9] The treatment consists in annealing the sample at 10 Torr oxygen pressure (P_{O_2}) and a temperature corresponding to the desired oxygen content line in the P_{O_2}-T phase diagram.[10] After this, the sample is slowly cooled through the desired oxygen content line on the P_{O_2}-T phase diagram. This controlled cooling is followed by a rapid quench to RT from ~350 °C. In this fashion, high T_c films of desired oxygen stoichiometry with narrow transition widths are reproducibly obtained.[9] The laser irradiation was done using an Ar ion laser with a series of lines in the 454.5 nm $\leqslant\lambda\leqslant$ 514.5 nm range and a total power of 6 W. The samples were immersed in liquid nitrogen during irradia-

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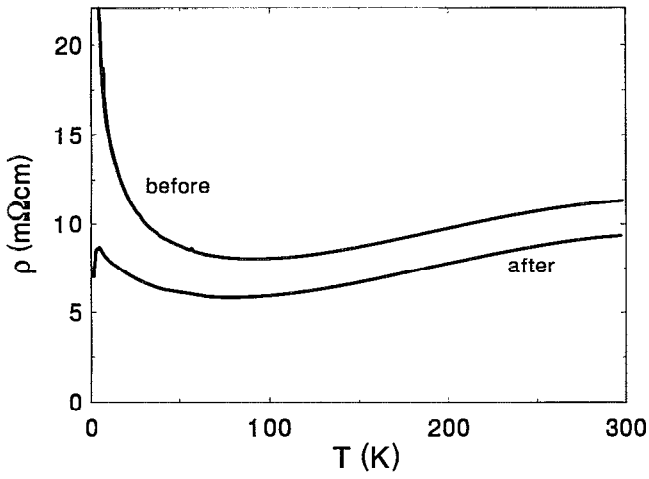

FIG. 1. Electrical resistivity as a function of temperature before and immediately after 12 h laser illumination of a 200-nm-thick $GdBa_2Cu_3O_x$ film.

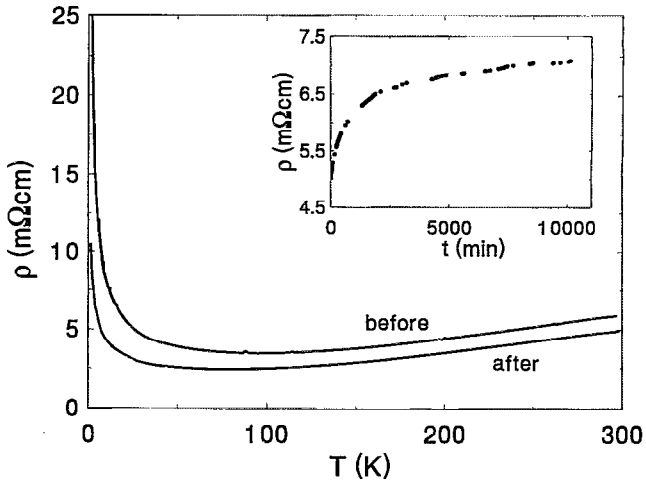

FIG. 3. Electrical resistivity as a function of temperature, T, before and immediately after laser illumination for the $T_c = 0$ K film. The inset shows the relaxation of the resistivity at RT.

tion and the change in resistivity was measured *in situ* during photoexcitation. Contacts to the sample were made with sputtered Ag electrodes. The amount of heating during irradiation was minimal, as indicated by the absence of abrupt changes in the sample resistivity after the laser was turned off. After the illumination the relaxation of the resistance was measured at RT.

Figure 2 shows the time evolution under laser illumination of the 77 K normal state resistivity for three 100-nm-thick YBCO samples on MgO; one with a nominal oxygen stoichiometry[11] $x = 6.5$ and no detectable T_c down to 1.5 K (identified as $T_c = 0$ K), one with $x = 6.55$ and a zero-resistance critical temperature $T_c = 2$ K, and one with $x = 6.6$ and $T_c = 25$ K. All samples show initially a relatively fast decrease of the resistivity in 10–20 min, followed by a saturation plateau. Notice the correlation between the oxygen content and the relative change in the resistivity; i.e., the more "metallic" the sample, the smaller the relative change in the resistivity. Figure 3 shows the temperature dependent resistivity of the YBCO insulating sample ($T_c = 0$), before and after irradiation. The resistivity decreases throughout the entire temperature range, the change being an almost temperature independent shift above ~50 K. The inset shows the relaxation at RT after the illumination. Notice the difference in time scale with the excitation shown in Fig. 2. Figure 4 shows the change in the resistivity of the $T_c = 2$ K sample. Again, the shift in the resistivity is a parallel shift above ~50 K and a reduction of the "semiconducting like" behavior towards a "metallic" temperature dependence below ~50 K. The inset shows in an expanded scale the superconducting transition before and immediately after illumination. The effect on a sample which does not show an upturn in the resistivity is shown in Fig. 5. Again, the resistivity shift is almost constant above ~40 K. As indicated in the inset, the T_c is enhanced by almost 5 K after illumination. Notice that the superconducting transition shifts in a uniform way to higher temperatures, as already shown for the $T_c = 2$ K

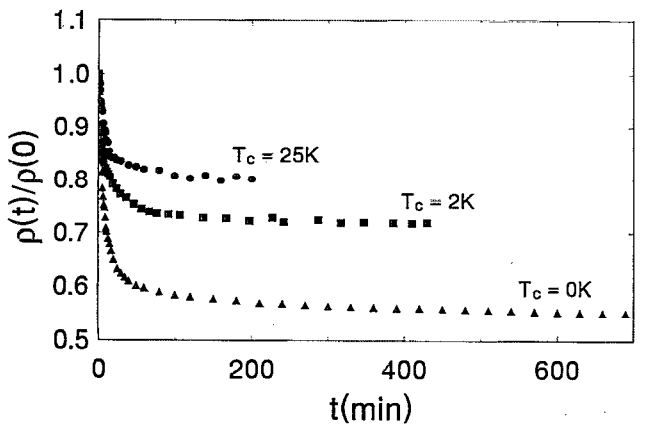

FIG. 2. Normalized electrical resistivity, $\rho(t)/\rho(0)$, at 77 K as a function of time, t, during laser illumination for three 100-nm-thick $YBa_2Cu_3O_x$ films with nominal oxygen stoichiometries: ▲, $x = 6.5$; ■, $x = 6.55$; ●, $x = 6.6$.

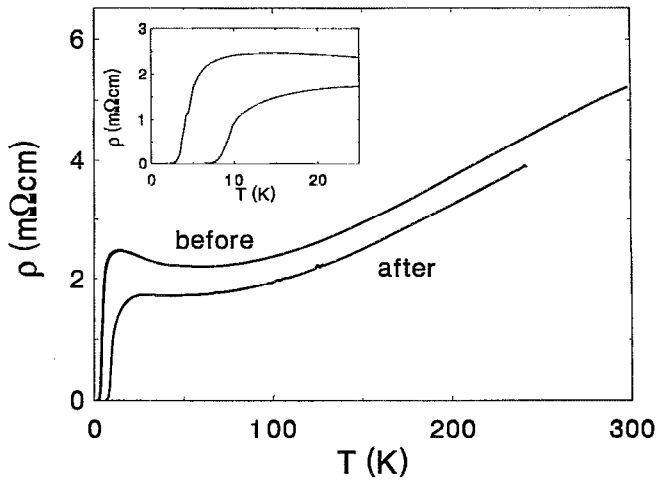

FIG. 4. Electrical resistivity as a function of temperature, T, before and immediately after laser illumination for the $T_c = 2$ K film. The inset shows the region near T_c in an expanded scale.

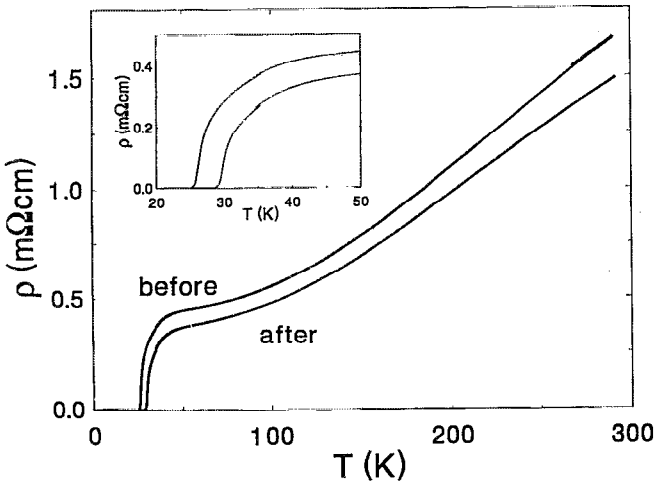

FIG. 5. Electrical resistivity as a function of temperature, T, before and immediately after laser illumination for the $T_c = 25$ K film. The inset shows the region near T_c in an expanded scale.

sample (inset Fig. 4). *Clearly, in both cases the superconducting transition is enhanced to higher temperatures, with the change being well outside the transition width and consequently independent of the T_c definition.* After 6 days in air at room temperature, T_c relaxes back to its original value within less than 1 K. However, the magnitude of the resistivity relaxes to a higher value than its initial value. After this relaxation, a second photoexcitation of the same film produces an identical T_c enhancement.

The ability to enhance the superconducting properties in a reproducible way opens up the possibility of preparing *in situ* weak links at low temperatures with desired properties. This could be accomplished in a more convenient fashion through the use of *in situ* solid-state lasers to maintain the desired properties for long periods of time.

The mechanism by which the enhancement of superconductivity occurs is not understood at the present time. One possible reason is that the laser irradiation changes the Josephson coupling between the superconducting grains, under the assumption that superconductivity in the films is granular in character. However, the fact that even a sample with a relatively high T_c and no upturn in the resistivity (Fig. 5) changes substantially, makes this explanation seem unlikely. Another possibility is that the changes are due to charge transfer induced by oxygen ordering[12] in the O1–O5 plane.[13] This requires photoexcitation to induce oxygen ordering in some fashion. What makes this explanation attractive is the fact that the laser photon energy ($\sim$2.2 eV) is of the same order as the chain oxygen (O1) activation energy[14] ($\sim$1.2 eV) and that the relaxation times for oxygen ordering[12] are very similar to the ones measured here. However, the mechanism by which laser irradiation would induce ordering, whereas temperature induces disordering, as indicated by the room temperature relaxation, is not clear. Of course, the possible formation of long-lived metastable oxygen complexes which change the hole doping of the CuO_2 planes, cannot be ruled out. A third possibility, the photogeneration of additional mobile holes in the CuO_2 planes, is not easily understood, due to the many orders of magnitude difference between the observed relaxation times and typical inelastic electron collision times.[15] However, it could be argued that this relaxation rate is enhanced due to anisotropy and low carrier density effects[16] or due to trapping effects,[3] the latter being less likely to occur in the "metallic" sample. Clearly, more theoretical work is needed to clarify this issue.

In summary, we have observed sizable enhancements in the superconducting properties of RBCO thin films by photoexcitation. This experimental discovery opens up the possibility of reproducible production of weak links through *in situ* tuning of superconducting properties. The microscopic origin for the enhancement is not clear at the present time and is currently under investigation.

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