Non-exponential optical modification of the electrical properties in oxygen-deficient YBa$_2$Cu$_3$O$_x$

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Abstract. We present studies of the time dependent electrical resistivity of YBa$_2$Cu$_3$O$_x$ during visible light excitation (photodoping). Illumination with a green semiconductor laser changes the resistivity in a non-exponential way, which is well described by a stretched-exponential function with parameters $\beta$ (dispersion parameter) and $\tau$ (time constant). $\beta$ showed an unusual behaviour: at temperatures below 220 K $\beta$ was about 0.3 to 0.4, but increased near room temperature to 0.5 to 0.6. For comparison with results obtained by red-laser excitation we discuss the dependence of the resistivity on the cumulative photon dose, thereby introducing a “photon dose constant” $N_e$ which corresponds to $\tau$. The results indicate that green-light excitation is somewhat more effective than red-light excitation at low temperatures.

1. Introduction
Persistent photoconductivity (PPC) in partially oxygen-deficient YBa$_2$Cu$_3$O$_x$ was extensively studied (see e.g. Refs. [1] and [2] and references therein). It was established that the photo-induced conductivity enhancement shows relaxation only above $\approx$ 250 K and significantly depends on the wavelength of the exciting radiation and on the temperature $T_{dop}$ at which the sample illumination is carried out. Most studies of the wavelength dependence, however, were performed using the slope of the resistance $R$ vs. illumination time $t$ dependence at small photon doses, i.e. the region where $R(t)$ is nearly linear, as the only measure for the photodoping efficiency. For potential applications it is far more interesting to have detailed knowledge of the relation between $R$ and the cumulative photon dose for various temperatures and excitation energies.

The present work adresses the above subject. We started a detailed study of the time dependent $R$ in oxygen-deficient YBa$_2$Cu$_3$O$_x$ during light excitation at various temperatures, applying photon doses up to $10^{23}$cm$^{-2}$ and using several different lasers as the light source. Here we present the results and conclusions from a first series of experiments in which a green semiconductor laser was used for the illumination.

2. Experimental techniques
The experimental methods in this work were essentially the same as in Ref. [3] (see also the references therein). We used a thin film 60-K YBa$_2$Cu$_3$O$_x$ ($x \approx 6.6$) prepared by pulsed-laser deposition on a “wedged” (off-axis) SrTiO$_3$ substrate with a tilt angle of 10°. During the
Figure 1. The dependence of the normalised resistivity $R(N)/R(0)$ of YBa$_2$Cu$_3$O$_x$ on the cumulative photon dose $N$ during excitation with the green-light semiconductor laser.

deposition, the substrate was kept at 760 °C in an oxygen atmosphere of 0.7 mbar. After deposition, the film was annealed *in situ* for 100 min without changing the temperature and oxygen pressure. $T_c$ of the non-illuminated sample was about 58 K. A cross-shaped test structure was fabricated by means of photo-lithography and wet chemical etching. Samples such like this can be used to study the anisotropic properties of YBa$_2$Cu$_3$O$_x$ [4]. The present work, however, deals only with the in-plane resistivity of the sample.

During the photodoping experiments, the sample was kept in a temperature controlled closed-cycle refrigerator equipped with a Suprasil window allowing for the illumination of the sample. The electrical resistivity was measured with a standard 4-point method using a highly-stable ac current source and lock-in technique. The sample was illuminated for about 24 hours with a green semiconductor laser ($\lambda = 532$ nm, $\hbar \omega = 2.3$ eV) . The light intensity on the sample surface was about 0.6 W/cm$^2$.

3. Results and discussion

Figure 1 shows the dependence of the normalised resistance of the sample as function of the cumulative photon dose $N = It/\hbar \omega$ ($I$ is the light intensity and $t$ is the illumination time) for 5 different temperatures. The observed variation of the photodoping effect is in good agreement with previous results obtained by white-light photodoping [2]: it is large at low temperatures, becomes smaller in the intermediate temperature range (120 K to 220 K) and is large again at high temperatures. However, there is one significant difference: in the white-light experiments, the resistance reduction was largest at high temperatures ($T > 260$ K), whereas in figure 1 it is larger at 70 K than at 270 K. Obviously, the effect of photodoping varies with the temperature $T_{dop}$, and this variation also depends on the photon energy of the exciting radiation.

The curves in figure 1 were fitted to the stretched-exponential function $R(N) = R_{min} + [R(0) - R_{min}] \exp[-(N/N_e)^\beta]$, where $N_e$ is a “photon dose constant” which corresponds to $\tau$, and $R_{min}$ is the limit of $R$ for $N \to \infty$. The theoretical values were in excellent agreement with the data. Hence, for more clarity, we have not plotted them in figure 1. The fitted parameters, as given in table 1, show that $\beta$ is about 0.3 to 0.4 at low and intermediate temperatures, but...
increases towards 0.5 - 0.55 at high temperatures.

The temperature dependence of $\beta$ is completely different from that observed in PPC relaxation experiments [1], where $\beta$ varied linearly between 0.5 and 0.7. Our findings suggest an interpretation based on the microscopic theory of $\beta$ by J.C. Phillips [5]. Within this theory, $\beta$ is determined by the effective dimensionality of the configuration space in which the trapping of photo-excited charge carriers takes place. Phillips derived two “magic values” of $\beta$: 0.43 and 0.60, valid for long-range and short-range Coulomb interactions, respectively. The results in table 1 are close to those magic values and may indicate a crossover between the two types of Coulomb interactions occurring at temperatures above 200 K.

**Table 1.** Parameters of the stretched-exponential fits to the data in figure 1. The accuracy of the parameter values is about 5 %.

| T(K) | $\beta$ | $N_e (10^{22} \text{cm}^{-2})$ | $R_{\text{min}}/R(0)$ |
|------|---------|-------------------------------|-----------------------|
| 70   | 0.44    | 1.96                          | 0.895                 |
| 120  | 0.32    | 1.20                          | 0.943                 |
| 170  | 0.44    | 1.10                          | 0.976                 |
| 220  | 0.54    | 6.21                          | 0.943                 |
| 270  | 0.51    | 4.15                          | 0.912                 |

The PPC effect in oxygen-deficient YBa$_2$Cu$_3$O$_x$ is attributed to the trapping of photo-excited electrons in the CuO-chain layer of the unit cell. In a first step, electron-hole pairs are created within the CuO$_2$ planes and (small) fraction of the electrons are transferred to the chain layer, where they are trapped in chain defects. For the trapping the electron has to find an appropriate defect state within its life time. This is the point were the model by Phillips may become relevant. The nature of the interaction between the electron and the defect will determine the fraction of photo-excited electrons that is trapped.

![Figure 2](image.png)

**Figure 2.** The $T_c$ enhancement caused by the photodoping experiments in figure 1 plotted as a function of $T_{\text{dop}}$. The line is a guide to the eye.

After each of the photodoping experiments we measured the photo-induced enhancement $\Delta T_c$ of the superconducting transition temperature $T_c$. $\Delta T_c$, plotted in figure 2 versus $T_{\text{dop}}$, is 0.3 K to 0.4 K at low and intermediate temperatures, but rises to about 0.7 K at 270 K. These results are in excellent agreement with our previous results, obtained in white-light photodoping
experiments [2]. The value of $T_c$ is determined by the normal-state free carrier concentration. Therefore, the comparison between figures 1 and 2 indicates that the large resistance reduction at 70 K is caused by a photo-induced enhancement of the carrier concentration and by a significant increase of the carrier mobility, in agreement with the previous studies [2].

For comparison we performed an additional photodoping experiment, with a Helium-Neon laser ($I = 1 \text{ W/cm}^2$) as the light source and adapting the illumination time appropriately to achieve the same cumulative photon dose as with the green laser. The results are shown in figure 3 together with the 70 K measurement from figure 1.

![Figure 3. Comparison of the photodoping effect for He-Ne laser illumination (1) and the illumination with the green-light laser (2). Both measurements were taken at 70K.](image)

The stretched-exponential parameters for the He-Ne laser illumination were: $\beta = 0.47 \pm 0.03$, $N_e = (3.02 \pm 0.10) \times 10^{22} \text{cm}^{-2}$ and $R_{\text{min}}/R(0) = 0.906 \pm 0.005$. Figure 3 suggests that, at 70 K, the effect of photodoping is somewhat smaller for the He-Ne laser illumination. However, this assessment appears questionable if additional data are taken into account. $\Delta T_c$ with the He-Ne laser was 0.37 K, which is slightly larger than with the green-light laser (see figure 2). Also, the values of $R_{\text{min}}/R(0)$ were not significantly different, considering the accuracy of the parameter values. Obviously, it can be misleading to measure the efficiency of photodoping only by the slope at small photon doses.

The detailed knowledge of $R(N)$ in a wide temperature range and for various photon energies may also be of technological interest in the future. For example, an oxygen-deficient thin film of YBa$_2$Cu$_3$O$_x$ is able to keep the information about light-illumination doses for a long time after the cessation of the illumination. It is conceivable that such photon dose memory devices could be useful for research as well as technical applications.

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