Photoinduced Superconducting Nanowires in GdBa$_2$Cu$_3$O$_{6.5}$ films.

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Abstract

We report the fabrication of high $T_c$ superconducting wires by photodoping a GdBa$_2$Cu$_3$O$_{6.5}$ thin film. An optical near-field probe was used to locally excite carriers in the system at room temperature. Trapping of the photogenerated electrons define a confining potential for the conducting holes in the CuO planes. Spatially resolved reflectance measurements show the photogenerated nanowires to be $\sim 250$ nm wide. Electron diffusion, before electron capture, is believed to be responsible for the observed width of the wires.
One of the most important prospects with high temperature superconductors is their application in electronic circuits. Therefore the capability to define narrow lines and small Josephson junctions is critical for the technological development of these materials. Many applications would benefit from submicron feature sizes, not attainable with conventional patterning and etching. An alternative method to define superconducting wires is suggested by the persistent photoconductivity (PPC) and persistent photoinduced superconductivity (PISC) effects observed in oxygen-deficient $\text{RBa}_2\text{Cu}_3\text{O}_{6+x}$ (RBCO) superconductors. It has been shown that under visible and ultraviolet illumination photodoping closely resembles the more common oxygen order self doping in RBCO, increasing the carrier density. The relaxation time of photogenerated carriers is several hours at room temperature, and becomes persistent if the sample is kept at temperatures below 100 K. Recent experiments have shown that electron detrapping involves oxygen ion movement, suggesting that captured electrons will be localized on a scale of order of the dimensions of the unit cell. Since all of the previous experiments were carried out with broad area illumination, this small localization length has not been experimentally exploited.

In this Letter we show that photocarriers can be induced and confined on a scale of 250 nm by illuminating the sample with a near-field scanning optical microscope (NSOM) probe. A $c$-axis oriented, 180 nm thick $\text{GdBa}_2\text{Cu}_3\text{O}_{6+x}$ (GBCO) thin film was grown on a (100) MgO substrate by dc magnetron sputtering, as described elsewhere. The as-grown film shows linear temperature dependence for the DC resistivity $T_c$ of 89.4 K, with a transition width of 1 K, as measured by AC susceptibility measurements. The oxygen content of the sample was adjusted to $x=0.5$, and the sample was patterned by conventional photolithography and wet etching into a four probe geometry.

The sample was mounted on a continuous-flow He cryostat and photodoped with a 100 nm diameter probe of a room temperature NSOM. The tip-to-sample separation was controlled in the 5-10 nm range by means of a quartz mechanical oscillator feedback system. Light from either a 10 mW, $\lambda = 632.8$ nm HeNe laser or a 1 mW, $\lambda = 1.55 \mu m$ InGaAsP
laser diode was coupled into the NSOM probe by means of a 2x2 optical fiber coupler. During photogeneration the sample was illuminated only by the HeNe laser. Reflectance changes were measured by illuminating the sample through the NSOM probe (with only the 1.55 µm laser light coupled into it) and collecting the reflected light with a conventional microscope objective. Photon fluxes per unit time in the near field were estimated to be \( Q(632.8\text{nm}) \sim 3.5 \times 10^{21} \text{photons cm}^{-2}\text{sec}^{-1} \) and \( Q(1.55\mu\text{m}) \sim 2.5 \times 10^{21} \text{photons cm}^{-2}\text{sec}^{-1} \). For resistance measurements the NSOM head was detached, the cryostat closed and pumped to 10^{-6} Torr and the sample cooled to 120 K. It took less than 20 min between the end of photogeneration at room temperature and cooling the sample to 120 K.

The main result of this paper is presented in Fig. 1. The NSOM reflectance scan, obtained after illuminating on a 1 µm side square for \( t = 2000 \text{ sec} \), demonstrates \( w \sim 250 \text{ nm} \) confinement of photogenerated carriers. The scan took 10 min to acquire, and was measured from bottom to top in Fig. 1. The different contrast between the bottom and top sides of the square is due to the ongoing photocarrier recombination.

The time dependence of photocarrier recombination was further investigated at the point marked by an arrow in Fig. 1. Raster scans across the nanowire were averaged 100 times during a time span of 10 sec and the process was repeated every 30 sec. The results, shown in Fig. 2a, are consistent with the current understanding of photocarrier generation in RBCO. A trapped electron has a probability \( P_i \propto \exp(-\Delta_s/k_BT) \) to become ionized where \( \Delta_s \sim 1 \text{ eV} \). Room temperature recombination of electrons with holes from the CuO planes decreases the available number of free carriers and, hence, the optical conductivity at 1.55 µm. Fig. 2b shows that the integrated intensity decreases at a rate slower than exponential, as has been observed in wide area PPC experiments. Although the dynamical range is too small to draw strong conclusions, the fact that the decay is not a pure exponential suggests that even an area of \((250 \text{ nm})^2\) presents a distribution of \( \Delta_s \). The most important information about the time evolution is contained in Fig. 2c, which shows the time dependence of the full-width half-maximum (FWHM) of the line profile intensity. The FWHM is larger than the diameter of the NSOM probe and increases only slightly more
than the experimental error. As discussed later, the initial width is associated with the diffusion of photocarriers before electron capture, and the small observed increase in width could be associated with a much slower diffusion of trapped electrons.

To further investigate the properties of photogenerated carriers we have studied the low temperature resistance of photogenerated wires. A 20 µm long wire was written with the NSOM probe for 10 hours. After cooling the system to 100 K the rest of the sample was photoexcited by means of a focused HeNe laser beam. This last process was continued until no further change was observed in the DC resistance. The low temperature resistance of the photoexcited pattern is shown in Fig. 3a. The drop in the curve, at T ≈ 29 K, corresponds to the superconducting transition of the region photoexcited by the focused HeNe laser. At T ≈ 10 K an additional three orders of magnitude drop is observed in the resistance associated with the superconducting transition in the NSOM-induced wire. A V-I characteristic measured at 4 K confirms this result, as shown in Fig. 3b. For comparison, we also show the V-I characteristics of the photogenerated wire measured at 20 K as well as the result expected for a copper wire of similar dimensions at room temperature. Considering that our voltage noise was 70 nV, dissipation in the sample at 4 K and I ≈ 9 µA is more than three orders of magnitude smaller than in copper, and more than four orders of magnitude smaller than the sample at 20 K. The critical current, defined as the value where the voltage is larger than the noise, gives I_c ≈ 9 µA. This gives a critical current density J_c ≈ 10^4 A/cm^2, comparable with J_c for oxygen depleted samples with similar T_c.

The most puzzling feature of the data arises from the difference between the size of the NSOM tip and the width of the superconducting wire. Three main diffusive processes may be responsible for the extra width: Diffusion of the e-h pairs before electron trapping, diffusion of the trapped electrons, or diffusion of the conducting holes in the CuO planes. The extra holes pumped into the CuO planes move in the potential well defined by the trapped electrons, and they can not fluctuate more than a screening length, l_s. The density of photogenerated carriers was estimated to be n_e ≈ 10^{20} cm^{-3}, leading to a Thomas-Fermi screening length l_s ≈ 1 nm.
The results of Fig. 2c show that the other two processes are both present. Although $e-h$ diffusion before electron capture appears to account for most of the difference between probe and wire size, trapped electrons remain marginally mobile, as evidenced by the upward curvature observed in Fig. 2c. Thus localized electrons can hop randomly between trapping centers, similarly to the motion of carriers in semiconductor impurity bands. The hopping rate will then be given by

$$\tau^{-1} = \nu_{ph} \exp \left(-\frac{\Delta_s}{k_B T}\right),$$

(1)

where $\nu_{ph} \sim 10^{13}$ Hz is a typical phonon frequency. The two dimensional diffusion distance after 1000 sec is

$$\langle d^2_i \rangle = (30 \text{ nm})^2 = 4Nl^2 \sim 4 \frac{1000 \text{ sec}}{\tau} d_o^2,$$

(2)

where $N$ is the number of hops and $l$ is the hopping distance, which is taken as the separation between trapping centers $d_o \sim 1 \text{ nm}$, and 30 nm is the observed increase of the width at room temperature.

From Eq. (1) and (2), we get $\Delta_s \sim 850$ meV. This value is comparable to the excitation energy measured by PPC decay experiments [3,7] suggesting a common source for both processes. Since the captured electrons become thermally detrapped the $e-h$ pairs can recombine. The recombination time was estimated to be $\tau_R \sim 10^{-9} \text{ sec}$, [12] so that a trapping time $\tau_t \sim 4 \times 10^{-12} \text{ sec}$ is implied.

For technological applications a better understanding of the diffusion processes may allow control of the width of photoinduced wires. We believe that $e-h$ plasma diffusion is the dominant mechanism for determining the initial width of the wire, $d_i \sim 200 \text{ nm}$. Considering both hot electron diffusion and thermalized diffusion, we can express

$$\langle d^2_i \rangle = 4(v_{th}l_{th}\tau_t + v_B l_h \tau_h),$$

(3)

where $v_B(v_{th})$ is the velocity of the hot (thermal) electrons, $l_h(l_{th})$ their mean free paths, and $\tau_h$ is the time required for the electronic system to thermalize. The observed width
cannot be explained with the first term alone since the low thermal velocities of electrons 
\(v_{th} \sim 10^6 \text{ cm sec}^{-1}\) would require unreasonably large values for \(l_{th}\). Band velocities are in the 
\(10^8 \text{ cm sec}^{-1}\) range, however. For the hot carrier process to play a significant role \(\tau_h \sim 10^{-12} \text{ sec}\) is required. Although the thermalization time has been estimated to be \(10^{-13} \text{ sec}\), the agreement is not unreasonable in view of the uncertainties involved in the estimates. However, these considerations suggest that diffusion may occur in a more complicated fashion 

than represented by Eq. 3. Further studies of the processes involved are under way and will be published elsewhere.

In summary, a high \(T_c\) nanowire has been realized for the first time using NSOM photodoping. Photoholes transferred to the \(\text{CuO}\) planes move in a potential well defined by trapped electrons. The high spatial resolution of the NSOM probe provides new information about the physical phenomena and the characteristic parameters involved in the capture of photogenerated electrons. The nanowires, which width is determined by hot carrier diffusion, present enhanced optical conductivity and superconducting properties similar to those observed in oxygen doped materials. NSOM writing of high \(T_c\) superconducting wires opens new avenues towards the fabrication of superconducting devices.

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FIGURES

FIG. 1. Reflectance of the photogenerated wire measured with the NSOM at 1.55 \( \mu \text{m} \). The scanning area is \( 2 \times 2 \mu \text{m}^2 \). Maximum contrast is about 10%.

FIG. 2. Time evolution of the reflectance measured for raster scans across the wire. (a) Measured signal at 30 sec intervals. (b) Integrated intensity for each curve of (a). The dotted line represents a pure exponential behavior. (c) FWHM of the data of (a).

FIG. 3. Transport measurements of a photoinduced wire. a.- D.C. resistance as a function of temperature. b.- V-I curves measured at (●) 4 K and (—) 20 K on the sample. (····)Dissipation expected for a Cu wire of similar dimensions at room temperature.
This figure "figure2.GIF" is available in "GIF" format from:

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(a) Int. (a.u.) vs. position (µm) vs. time (sec)

(b) ΔR (a.u.) vs. time (min)

(c) FWHM (nm) vs. time (sec)
(b) Sample at 20 K

V (µV)

I (µA)

Cu at 300 K

R (Ω)

0 10 20 30 40 50 60 70

0 1200 1400

0 200 400 600 800 1000 1200 1400

I = 1 µA

T (K)