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Microbridge on $YBa_2Cu_3O_{7-\delta}$ thin film patterned by reversible laser annealing

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Abstract

A superconducting microbridge (14 μm wide, 140 μm long) on an epitaxial $YBa_2Cu_3O_{7-\delta}$ thin film was patterned by using a laser writing technique. The technique is based on a local increase of oxygen diffusion velocity caused by the heat of a focussed laser beam. Using nitrogen as the ambient atmosphere a deoxygenated (semiconducting) line could be written. A reoxygenated (superconducting) line forming a microbridge could be patterned by scanning the laser beam in ambient oxygen atmosphere perpendicular to the first written line. Neither optical reflection micrographs nor scanning electron microscopy images have shown any physical damage of the film surface due to this fully planar patterning technique. The reversible properties of the laser writing technique were proved by electrical four-point DC measurements between 30 K and 300 K.

Keywords: Applications of high-T_c superconductors; Metal–insulator transition; Oxygen stoichiometry; Thin films; Laser treatment

The patterning technique of HTS thin films are a crucial point for the performance of electronic applications. Several techniques suitable for patterning of YBCO films such as wet chemical and plasma etching, ion milling and laser ablation have been developed. These are all non-planar patterning techniques which ablate material, making it difficult to produce multilayer HTS devices without undesirable step-like grain-boundary weak links in the upper layers. Photolithographic and wet-etching processes often lead to patterns with contaminated (T_c suppressed) surfaces and fuzzy edges which can severely degrade device performances [1].

The technique we used for fabricating a microbridge is based on the observation that the electrical and optical properties of $YBa_2Cu_3O_{7-\delta}$ are very sensitive to its oxygen content. In the orthorhombic phase (i.e. $0 < \delta < 0.6$) $YBa_2Cu_3O_{7-\delta}$ behaves like a metallic conductor as regards the electronic transport properties, and below 92 K it becomes superconducting. The tetragonal phase of $YBa_2Cu_3O_{7-\delta}$ (i.e. $0.6 < \delta < 1$) behaves like a semiconductor. The oxygen content can easily be changed by focussed laser heating of the material, thereby causing locally an increase of the oxygen-diffusion velocity in the $YBa_2Cu_3O_{7-\delta}$ film [2–6]. The direction of the oxygen diffusion (i.e. into or out of the film) may be controlled by the application of either a reducing or oxidizing atmosphere. In this way oxygen-rich (superconducting) regions or depleted (semiconducting) ones can be patterned into the $YBa_2Cu_3O_{7-\delta}$ film by scanning the focussed laser beam across the surface. The process is planar, nonin-

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vasive, reversible, does not require a patterning mask, and does not contaminate the surface of the patterned film.

In this work we demonstrate the patterning of a 14 μm wide and 140 μm long superconducting YBCO microbridge by using the reversible properties of the laser-writing process. In contrast to Dye et al. [4] and Sobolewski et al. [6] deoxidizing the complete YBCO film by furnace annealing was not required before laser patterning the microbridge. Shen et al. patterned a microbridge in a similar way as we did [5]. In contrast to them no repeated laser scanning was required for patterning the microbridge in our case. Besides, we also measured the critical current in the superconducting state down to 69 K.

The superconducting $YBa_2Cu_3O_7$ films used here were prepared by reactive evaporation onto (100) MgO substrates (1×1 cm^2) [7]. The *c*-axis oriented, epitaxial films have typical onset transition temperatures around 87 K. The thickness of the films is ≈ 200 nm. By mechanically scratching parallel lines into the YBCO film with a diamond tip the film was separated in several electrically isolated stripes (1 mm × 1 cm) without contaminating the surface and possibly influencing the oxygen-diffusion properties of the sample [5]. In each separate stripe a microbridge was patterned by using an Ar ion cw laser (λ = 488 nm). During the patterning process either pure nitrogen or oxygen was flowing through the cell in which the YBCO film was mounted; the cell was kept at ambient temperature. The electrical properties of the written lines or the microbridges were determined by four-point DC resistance measurements in a closed-cycle refrigerator. For electrical contacts Ag pads were evaporated through a shadow mask on the YBCO film.

The microbridge was patterned in two steps. The first step was carried out in flowing nitrogen (1 atm). A 140 μm wide and 1 mm long deoxygenated line was written by scanning the focussed Gaussian laser beam (full width $w \approx 160$ μm at intensity $I/I_0 = 1/e$) across the film surface (laser power ≈ 4.5 W) dividing the mechanically scratched stripe in two halves. Each half was electrically brought in contact with two Ag pads.

Curve (a) in Fig. 1 shows a resistance versus temperature curve of a line written by scanning the laser beam with a velocity of 0.8 μm/s across the film surface. The increasing resistance with decreasing temperature points towards a value $\delta > 0.6$ in the written line. By choosing faster scan velocities of the laser beam it was possible to write deoxygenated but still superconducting lines with a T_c that was shifted to lower temperature. In a second step, in which pure oxygen was passed through the cell (1 atm), a superconducting oxygen-rich 14 μm wide microbridge was patterned by scanning the laser beam, now focussed to a 1/e width of 16 μm, perpendicular to the semiconducting line. In this case a scanning velocity 0.08 μm/s and a laser power 0.42 W led to good results. The resistance versus temperature data for the

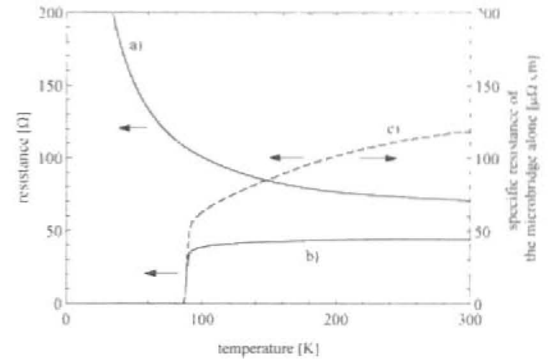

Fig. 1. (a) The resistance vs. temperature curve of a semiconducting line written in an YBCO film in nitrogen atmosphere. (b) The resistance vs. temperature curve for the semiconducting line shunted by a superconducting microbridge written in oxygen atmosphere. (c) The calculated resistance of the superconducting microbridge alone, without the contribution of the semiconducting parallel connection.

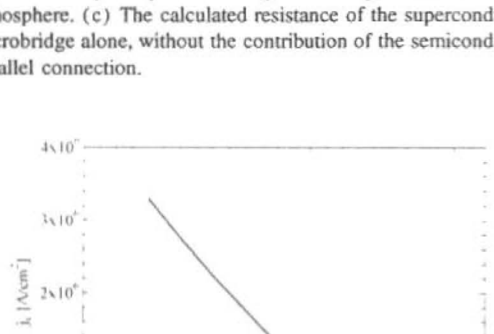

Fig. 2. Critical current density vs. temperature data for the laser patterned microbridge. The microbridge exhibited a critical current density of about 1.3×10^6 A/cm^2 at 77 K.

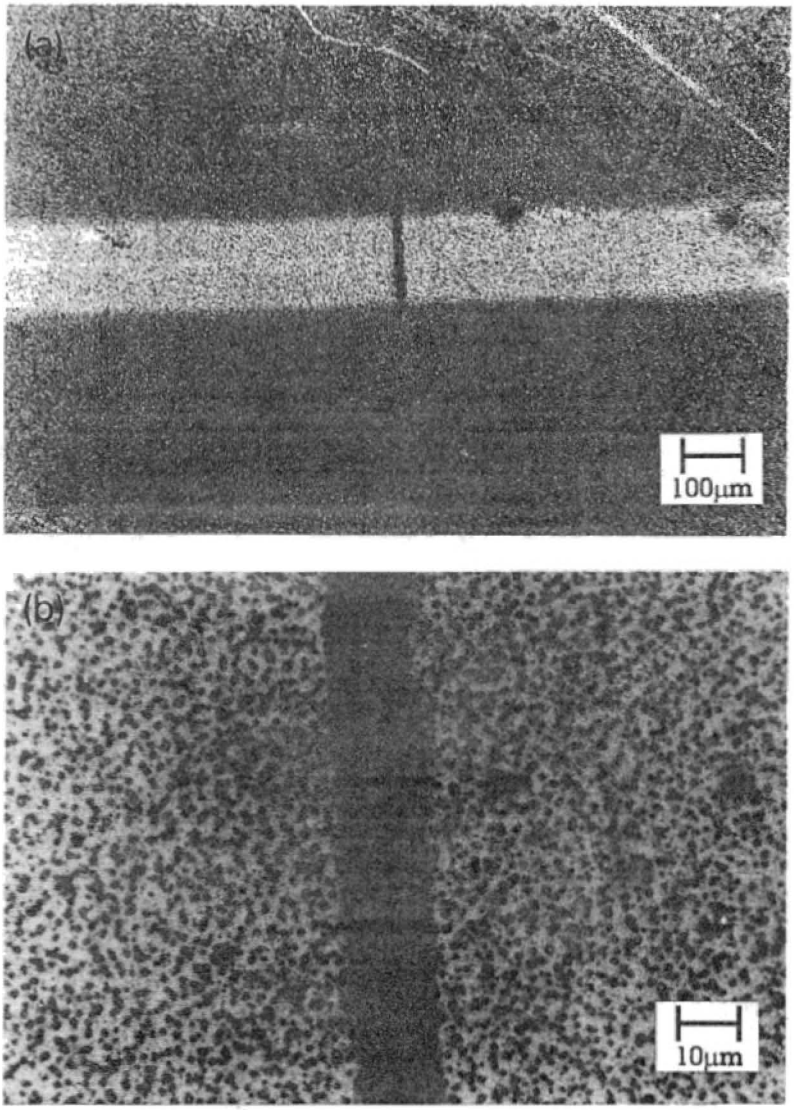

Fig. 3. (a) Optical reflection micrograph of the $YBa_2Cu_3O_{7-\delta}$ film after patterning the microbridge. The wide bright line (width ≈ 140 μm) was written in nitrogen atmosphere (1 atm). The dark thin bridge (width ≈ 14 μm) perpendicular to the nitrogen line was written in oxygen atmosphere (1 atm). (b) Enlargement of (a) showing the bridge in more detail.

semiconducting line shunted by a superconducting microbridge is shown in Fig. 1 curve (b). Curve (c) shows the calculated resistance of the superconducting microbridge alone without the contribution of the semiconducting line.

The laser-written microbridge exhibited a superconducting transition ΔT_c about 3 K wide (10–90%) with T_{c0} at 86 K, and a critical current density j_c of about 1.3×10^6 A/cm^2 at 77 K (Fig. 2). For the I_c measurements, we used a 1 μV criterion. The relatively wide superconducting transition and the nonlinear behaviour of the calculated ρ–T curve (c) in Fig. 1 points to oxygen-deficient regions or inhomogeneities in the microbridge. In the optical reflection micrograph (Fig. 3) the 140 μm deoxygenated line written in nitrogen is slightly brighter than the fully oxygenated surrounding film. The 14 μm wide oxygenated microbridge crossing the deoxygenated line has about the same brightness as the original unpatterned regions of the film. The contrast is caused by the oxygen dependence of the optical constants of $YBa_2Cu_3O_{7-\delta}$ [8], which yield a decreasing absorption coefficient in the visible part of the spectrum when $YBa_2Cu_3O_{7-\delta}$ is losing oxygen.

By contrast, scanning electron microscopy (SEM) images show no difference between oxygenated and deoxygenated lines. Neither in the optical reflection micrograph (Fig. 3) nor in the SEM image (not shown) any physical damage of the lines could be found. Only when higher laser powers were applied, signs of surface melting became apparent. Recent investigations of the laser patterned structures by means of a surface acoustic near field microscope (SNAM) demonstrate an unchanged surface topography but a significant increase of the work function at the deoxygenated parts of the YBCO film surface [9].

For estimating the temperature increase during the laser heating process we used a calculation which is based on a model of a two-layer system in which only the thin-film layer is absorbing [10,11]. For the thicker line written in nitrogen atmosphere as well as for the bridge written in oxygen atmosphere we calculate a surface temperature of approximatly 550 ± 100 °C during the writing process in the center of the laser focus. This value should be considered only as a rough estimate. First, because we used in our calculation constant values for the thermal conductivities which will not be true in reality due to the large temperature gradients in the two-layer system and a temperature dependence of the values. Secondly, there is an uncertainty in the thermal conductivity of the YBCO film. Investigations of the thermal conductivity of *c*-axis oriented YBCO films over a wider temperature range are still missing. Besides the published thermal boundary resistances of YBCO films on MgO (100) substrates differ widely [10].

In accordance with the calculated temperature profile caused by a Gaussian laser beam one can easily observe a much sharper transition between the dark superconducting and the brighter semiconducting regions in the case of the narrower 14 μm wide line compared to the 140 μm wide line.

The oxygen diffusion under our specific experimental conditions can be estimated by considering the film thickness and the time during which the YBCO film surface has to be heated to reach equilibrium, i.e. to become semiconducting or metallic, respectively. The time during which a certain point of the surface is heated is determined by the laser focus diameter and the scan velocity of the laser beam. As the relation between the focus diameter and the scan velocity was the same both in the case of writing the broad and the narrow line the heating time was also similar, namely $t \approx 200$ s. Yamamoto et al. [12] measured the time dependence of the conductivity of a *c*-axis film on MgO (off-axis magnetron sputtered) at a temperature of roughly 500°C for a sudden oxygen pressure change from 10 mTorr to 760 Torr. These authors obtained for their case of oxygenating an YBCO film a diffusion coefficient $D = 2.7 \times 10^{-12}\ \text{cm}^2/\text{s}$. In experiments with smaller pressure changes they found a smaller diffusion coefficient and no difference between in- and out-diffusion for temperatures $T > 500°\text{C}$. For oxygenating a laser ablated *c* oriented film on MgO Krauns et al. [13] determined in a similar way a coefficient $D(500°\text{C}) = 5.5 \times 10^{-13}\ \text{cm}^2/\text{s}$. The diffusion coefficients for the deoxygenation of the film were found to be one to two orders of magnitude smaller. To compare our required heating times with the measured diffusion coefficients in the literature we estimated the diffusion coefficient by a simple random walk model in the same way as Krauns et al. did, though it is possible that the diffusion under our experimental conditions is not even Fickian, i.e. that the flux is not linear in the concentration gradient [12]. In this simple model the coefficient D can be calculated from $\langle x^2 \rangle = 2Dt$. By setting $\sqrt{\langle x^2 \rangle}$ equal to the film thickness of 200 nm a diffusion coefficient $D \approx 1 \times 10^{-12}\ \text{cm}^2/\text{s}$ was estimated. This value lies between the values determined by Krauns et al. and Yamamoto et al. for oxygenating the film. Yet, it is not clear why we determine the same diffusion coefficient for in- and out-diffusion although Krauns et al. found for the deoxygenation a coefficient one to two orders of magnitude smaller than the one for the oxygenation. Both Yamamoto et al. and Krauns et al. observed a strong sensitivity of the oxygen diffusion to the surface structure of the used YBCO films. The dependence on the surface quality can be understood by the highly anisotropic diffusion in YBCO with diffusion coefficients in the *a*–*b* plane being about 10^4–10^6 times larger than in the *c* direction at elevated temperatures [14], so *c* oriented YBCO films with dense surface structure show a lower effective diffusion coefficient than films with lower surface quality, which provide a greater number of diffusion paths parallel to the *a*–*b* planes [12,13]. The scan velocities that were necessary in our experiments to reach a semiconducting behaviour or afterwards a superconducting one were slower than the velocities used by other groups [2–6]. In addition to different laser energy-densities and thermal conductivities of the film substrates also the surface microstructure of the YBCO film should have a strong influence on appropriate scan velocities.

In conclusion we have demonstrated that the laser annealing technique is suitable for patterning microbridges by just changing the ambient atmosphere from nitrogen to oxygen and using the reversibility of the process. A complete deoxidation of the YBCO film by furnace annealing was not required. Compared to conventional patterning techniques we see the major advantage of the used technique in its reversible properties. A further improvement of laser-written devices can be expected by in-situ controlling the electrical and optical properties during the patterning process.

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