Local modification of the thin $YBa_2Cu_3O_{7-y}$ microstrips by the voltage-biased atomic force microscope tip

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The atomic force microscope (AFM) tip biased at around −15 V is found to be capable of locally modifying the entire thickness of 40-nm-thick semiconducting or superconducting $YBa_2Cu_3O_{7-y}$ microstrips in air. We show, using combined electrical and AFM measurements, that the local regions underneath the surface of the semiconducting or superconducting $YBa_2Cu_3O_{7-y}$ microstrips are transformed into either nonconducting or nonsuperconducting regions, respectively, upon applying the negatively biased AFM tip. The conductance of the nonsuperconducting regions is also found to be comparable to that of the superconducting regions before modification at 298 K.
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One of the motivations to modify materials on smaller dimensions is to discover simple means for patterning novel quantum devices. One technique, widely known as a scanning probe-based local oxidation (SLO), has been demonstrated to be well suited for readily realizing quantum devices with new features.[1] SLO utilizes the voltage biased tip of an atomic force microscope (AFM) or a scanning tunneling microscope (STM) in ambient conditions to pattern dielectric barriers on thin conducting structures.

The high temperature superconducting $YBa_2Cu_3O_{7-y}$ (YBCO) is known to be highly reactive in the presence of water[2] and have an insulating or semiconducting surface layer after exposure to air.[3] Different examples and several models of surface modification of superconducting YBCO films with AFM or STM under ambient conditions have been reported.[4–6] In particular, Thomson *et al.* have demonstrated by using STM that lines of several hundred nm width that are raised as high as 100 nm from the surface of superconducting YBCO films can be controllably produced in the high field of the STM tip (>4 V sample bias).[4] Such raised lines on the YBCO surface resemble closely raised lines on the surface of other conducting films by SLO.[7] With the intention of investigating the modification mechanism and the hope of planar patterning YBCO junction devices, the present work has examined the possibilities offered by SLO to modify thin microstrips of the superconducting or semiconducting YBCO films.

Superconducting YBCO films 40 nm thick were grown by pulsed laser ablation onto MgO-(100) substrates. Photolithography and ion beam etching were used to pattern the superconducting YBCO films into 5 to 6-μm-wide strips, such as the ones shown in Fig. 3, connected to Au contact pads. The superconducting YBCO strips showed typical onset superconducting transition temperatures (onset T_c) of around 86 K in the film resistance versus temperature ($R-T$) curve. The superconducting YBCO strips became semiconducting after prolonged (12 weeks) exposure to air (with a relative humidity of about 60% at 298 K). The $R-T$ curve of the semiconducting YBCO strips was a rapidly increasing function of the temperature decrease without signs of the superconducting transition. The conductance of the semiconducting YBCO strips was about two orders of magnitude lower than that of the superconducting YBCO strips at 298 K.

Highly doped Si cantilevers (Park Scientific Instruments, Inc.) with 30-nm-thick Ti coating were used throughout this study. The AFM (Park Scientific Instruments, Inc., Auto Probe M5) was operated in a contact mode with a load force of 10.4 nN, and in air with a relative humidity of about 60% at 298 K. AFM modification experiments reported in this study were performed by scanning the AFM tip, which was biased to a negative voltage with respect to the strip surface, across the width of the YBCO strips at a rate of 8 μm/s.

To study the mechanism of the modification process, we monitored the time evolution of the strip conductance (G-t) while applying the negatively biased AFM tip across the YBCO strips. Knowing that G-t measurements made on other conducting microstrips provided a measure of the rate of the local oxidation,[8,9] we started our experiments with the following assumption: the conductance of YBCO strips will decrease noticeably if the local modification process alters the starting YBCO into nonconducting YBCO and progresses fairly deep into the film from the strip surface.

Figure 1(a) shows the G-t curve obtained while scanning the AFM tip across the semiconducting YBCO strip using −10 V tip bias. Here, the strip conductance decreases by an order of magnitude from 495 to 45 nS in 60 s of applying the tip bias. The current–voltage ($I-V$) curve obtained before applying the tip bias was linear but as shown in Fig. 2(a), it became nonlinear afterwards. Such nonlinear $I-V$ curve is reminiscent of metal-insulator-metal (MIM) tunnel diodes fabricated by SLO.[8,9] The surface topography [Fig. 3(a)] and the height profile [Fig. 3(e)] of the semiconducting strip obtained after applying the −10 V tip bias show that a broad line of about 2 μm width and 7 nm height is produced across the strip.

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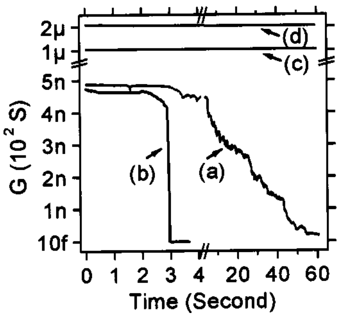

FIG. 1. The strip conductance vs modification time (G-t) curves at 298 K recorded while scanning the AFM tip at a rate of 8 μm/s: across the semiconducting YBCO strips using the tip bias of (a) -10 V or (b) -15 V; across the superconducting YBCO strips using the tip bias of (c) -10 V or (d) -15 V.

Figure 1(b) shows the G-t curve obtained while scanning the AFM tip across the semiconducting YBCO strip using -15 V tip bias. Initially, the conductance of the strip decreases slowly but it drops increasingly faster below the current detection limit of 10 pA within 4 s of applying the -15 V tip bias. The profile of the G-t curve in Fig. 1(b) resembles closely the profile of G-t curves that represent the current or voltage-induced local oxidation of thin metal films.[8,9] The $I-V$ curve obtained before applying the -15 V tip bias is linear and, as shown in Fig. 2(b), the $I-V$ curve obtained afterwards clearly confirms the complete electrical isolation. Applying the -15 V tip bias across the semiconducting strip also induced a relatively broad line [Fig. 3(b)] that is about 2 μm wide and raised about 20 nm high from the strip surface [Fig. 3(f)].

Since we do not find any signs of indentations in the AFM images of the modified semiconducting strips, we neglect reduction of electrical paths due to material removal as a cause for the conductance decrease observed in the G-t curves of Figs. 1(a) and 1(b). Rather, the volume expansion, the nonlinearity in the $I-V$ curve, and the profile of the G-t curve provide strong evidence that the conductance decrease is caused by reduction of electrical paths due to volume increase of nonconducting regions within the semiconducting YBCO strips by the modification process similar to SLO.

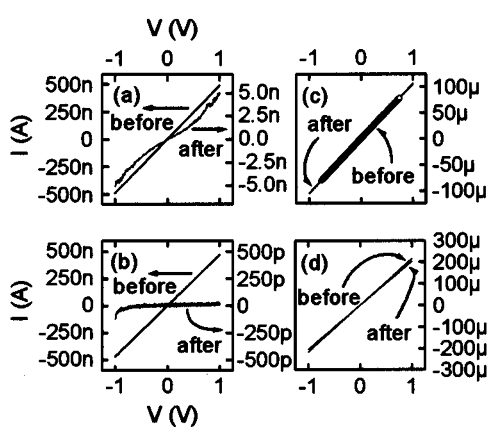

FIG. 2. Current–voltage ($I-V$) curves at 298 K: $I-V$ curves in (a), (b), (c), and (d) were obtained before and after applying the AFM modification processes of Figs. 1(a), 1(b), 1(c), and 1(d), respectively, across the modified strips shown in Figs. 3(a), 3(b), 3(c), and 3(d), respectively.

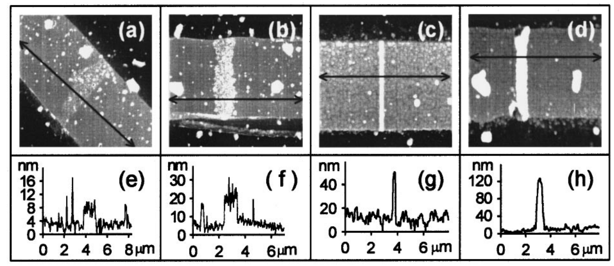

FIG. 3. AFM images of (a), (b), (c), and (d) were taken after applying the AFM modification processes of Figs. 1(a), 1(b), 1(c), and 1(d), respectively. The height profiles taken along the closed arrows indicated in (a), (b), (c), and (d) are shown in (e), (f), (g), and (h), respectively. The scanned area of (a) or (b) is 7 μm×7 μm. The scanned area of (c) or (d) is 8 μm×8 μm.

The G-t curve obtained while scanning the AFM tip across the superconducting YBCO strip using -10 V tip bias is shown in Fig. 1(c). Here, the conductance of the superconducting strip remains at constant level during the entire 60 s of applying the -10 V tip bias. $I-V$ curves obtained before and after applying the -10 V tip bias are linear and, as can be seen in Fig. 2(c), they precisely overlap. Even though applying the -10 V tip bias did not alter the conductance of the superconducting strip at 298 K, it produced a well-defined line [Fig. 3(c)] of about 250 nm width that is raised 35 nm high from the strip surface [Fig. 3(g)].

Figure 1(d) is an another G-t curve obtained while scanning the AFM tip across the superconducting YBCO strip using -15 V tip bias. In this case, the strip conductance decreases by about 3% from 209 to 203 μS in 60 s of applying the -15 V tip bias. $I-V$ curves obtained before and after applying the -15 V tip bias are linear and, as shown in Fig. 2(d), they are consistent with the conductance change in the G-t curve. Applying the -15 V tip bias gave rise to a line of increased size across the superconducting strip and, as shown in Fig. 3(d), it introduced indentations near the ends of the line. The height profile shown in Fig. 3(h) reveals that the line is about 700 nm wide and raised about 100 nm high from the strip surface. The further analysis of height profiles identified that the indentations penetrated as deep as about 40 nm into the film from the strip surface. Such indentations can reduce the cross section for electrical conduction which, in turn, is believed to be responsible for the small conductance drop shown in the G-t curve of Fig. 1(d).

To examine how deep the local modification progresses from the surface of superconducting strips and to verify the seemingly conducting nature of the modifications induced within the superconducting strips, we measured the $R-T$ characteristics of the modified superconducting strips featured in Figs. 3(c) and 3(d). As shown in the normalized $R-T$ curves of Fig. 4, the offset T_c is decreased from 65 K [Fig. 4(a)], 50 K [Fig. 4(b)], to below 10 K [Fig. 4(c)] with increasing the extent of modification by applying the tip bias from none (i.e., a unmodified superconducting YBCO strip), -10 V, to -15 V, respectively. We associate the degree of decreasing in the offset T_c with the degree of reduction in superconducting paths caused by volume increase of nonsuperconducting regions produced locally within the strips. The presence of nonzero resistivity even at 10 K, as shown in Fig. 4(c), suggests that the local modification does progress fairly deep into the film from the surface of the supercon-

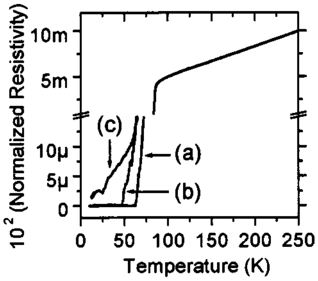

FIG. 4. Normalized resistivity $[R(T)/R(250\,\mathrm{K})]$ vs temperature (T) curves of: (a) an unmodified superconducting YBCO strip; (b) the modified superconducting YBCO strip shown in Fig. 3(c) using -10 V tip bias; (c) the modified superconducting YBCO strip shown in Fig. 3(d) using -15 V tip bias.

ducting strip. The linearly overlapped $R-T$ curves visible in the temperature range above the onset T_c provide additional evidence that the nonsuperconducting products produced within the superconducting strips are as conductive as the starting superconducting YBCO at temperatures above the onset T_c.

Similarly to the modification mechanism proposed by Bertsche *et al.*,[6] we think that, initially, barium ions on the surface sites react locally with ambient water and ambient carbon dioxide to form barium carbonate on the YBCO surface in the field of negatively biased AFM tip. Barium ions from the unmodified YBCO may be extracted onto the reacting surface by field-induced diffusion in such a way that the crystallites of barium carbonate on the surface grow in size. This may allow the transformation of local regions underneath the nonconducting crystallites into either nonconducting or nonsuperconducting YBCO, depending on the degree of the superconductivity of the starting YBCO. In agreement with our thought, Thomson, Moreland, and Roshko reported the inaccessibility of STM tunneling to raised lines formed on the surface of superconducting YBCO films, indicating that the raised lines are nonconducting.[4]

In summary, we have shown that the conductive AFM tip biased at -15 V is capable of locally modifying the entire thickness of 40-nm-thick YBCO microstrips in air. Electrical measurements revealed that the local regions underneath the surface of the semiconducting or superconducting microstrips are transformed into either nonconducting or nonsuperconducting regions, respectively, upon applying the voltage-biased AFM tip. The successful patterning of the MIM diode on the semiconducting YBCO strip provides evidence that the modification process identified in this study is promising for obtaining planar-type YBCO junction devices.

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