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Tailoring $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ surface Josephson junctions

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ABSTRACT

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO) single crystals are promising for high-temperature superconducting electronic devices with ultimate performance. Recently, tailoring superconductivity in ultra-thin BSCCO locally has received wide attention and interest. In this work, we focus on controlling the superconductivity of the uppermost CuO_2 double layer of the BSCCO crystal. Gold electrodes are deposited onto the freshly cleaved BSCCO surface by a high-vacuum *in situ* evaporation technique at room temperature, and then surface Josephson junctions (SJJs) between the two outermost superconducting CuO_2 double layers are fabricated as a probe to evaluate the superconductivity of the uppermost double layer. We find that the junction transition temperature T_c' as well as its critical current density j_c depend systematically and reproducibly on the deposition rate of the gold electrode. The electric properties of the SJJs, spanning the range from completely degraded to almost as good as the intrinsic Josephson junctions inside the bulk, systematically reveal the evolutionary process of superconductivity of the uppermost BSCCO layer, extending the range for practical applications.

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Superconducting electronic devices are important for a broad range of applications in quantum computation,^{1,2} remote sensing,^{3,4} secure communication,^{5,6} radio astronomy,^{7,8} microwave and millimeter wave technology,⁹ etc. However, most devices operate below the temperature of liquid helium. Some special applications like space communications are subject to costly and bulky cryocoolers. Therefore, there has been a continuous exploration of high-temperature superconducting electronics for more practical temperature ranges for decades.^{10–12}

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO) single crystals are highly promising for superconducting electronics due to easy cleavage, atomically smooth surfaces, and attainable ultra-thin superconducting flakes. BSCCO stacks have been intensively studied in the context of terahertz (THz) sources and detectors,^{13–21} and recently, the application of ultra-thin BSCCO layers creates great interest.^{12,22,23} In particular, superconducting nanowire single-photon detectors (SNSPDs) made of ultra-thin BSCCO layers exhibited excellent single-photon sensitivity at telecommunication wavelengths.^{24,25}

However, BSCCO is very sensitive to heat, air, and chemical solvents during the fabricating process, making the surface layer degraded or even insulating.^{26–28} Although ultra-thin BSCCO layers were usually covered by two-dimensional materials such as graphene to prevent degradation, their superconducting properties were still far from optimal,^{29–31} greatly affecting the reliability and stability of devices. The working temperature of ultra-thin BSCCO SNSPDs was also restricted to values below 20–25 K by degradation, which was far below the critical temperature of bulk crystals ($T_c \approx 90$ K).^{24,25} As a result, reducing or even completely avoiding the degradation of ultra-thin BSCCO layers is believed to be the inevitable key issue for practical devices.

Since the transport properties of a Josephson junction strongly depend on the properties of its two superconducting electrodes, the surface Josephson junctions (SJJs), formed between the two uppermost CuO_2 double layers of BSCCO acting as superconducting electrodes and the BiO and SrO layers in between acting as a barrier,³² are an excellent probe to evaluate the degradation degree of the uppermost BSCCO layer. In previous studies, the critical temperature of the SJJs

(T_c') was brought up to 80 K by an *in situ* cleavage technique,³³ which was pretty close to T_c . However, the cleavage needed to be done at liquid nitrogen temperature, which complicated the manufacturing process and made it costly.

In this Letter, a convenient and reproducible technique to create SJJ's with a well-defined degree of degradation is presented, which is a high-vacuum *in situ* evaporation technique at room temperature. By controlling the evaporation rate of the gold electrode, we are able to tailor T_c' reproducibly from completely degraded to as high as T_c of the bulk. Correspondingly, the (nominal) low-temperature critical current densities j_c range between 8 and 1260 A/cm². Note the latter value is almost the same as the j_c of the inner intrinsic Josephson junctions (IJJs), demonstrating that one can obtain a "perfect" superconducting layer on the BSCCO surface with negligible degradation. In addition, thanks to the obtained broad range of junction parameters, there is the chance to systematically study the transport properties of these SJJ's to better understand the electromagnetic coupling between the BSCCO layers. Our results show that the superconductivity of the topmost CuO₂ double layer can be precisely tuned, extending the range of potential applications of high- T_c BSCCO crystals.

The BSCCO single crystals are grown by a floating-zone technique and are used *as grown* to pattern the SJJ's. The sample structure is shown in Fig. 1(a) with an equilateral-triangle gold electrode (right)

in direct contact with the surface of an equilateral-triangle BSCCO stack. Typical optical images of the SJJ's are shown in the inset of Fig. 1(b). The 200-nm-thick gold electrodes in contact with the BSCCO stack are prepared by a high-vacuum *in situ* evaporation technique at room temperature allowing for deposition of the gold film with a well-defined deposition rate r immediately after cleavage. A series of samples is prepared by changing the gold deposition rates r between 6 and 26.9 nm/s. For clarity, we describe the process in detail. A small piece of the BSCCO crystal is placed in a high-vacuum evaporator, which is pumped to less than 2×10^{-8} Torr by a Pfeiffer TC 400 turbo pump. The BSCCO crystal is connected to the shutter of the evaporator by a piece of scotch tape for *in situ* cleavage. Before cleavage, gold particles in a tungsten boat are warmed up to a stable high-temperature state matching a desired deposition rate r , which is controlled by the evaporator amperage. We vary amperage in steps of 10 A from 131.5 to 171.5 A. The deposition rate has been calibrated by a step profiler before the fabrication process, so that the relation between r and the evaporator amperage is known. Since the BSCCO crystal is connected to the shutter, the gold deposition starts in the same moment when cleavage ends.

The bare part of the BSCCO stack is deteriorated and seemingly becomes insulating. Thus, underneath the contacting electrodes, the current needs to flow along the c -axis before in-plane transport occurs,

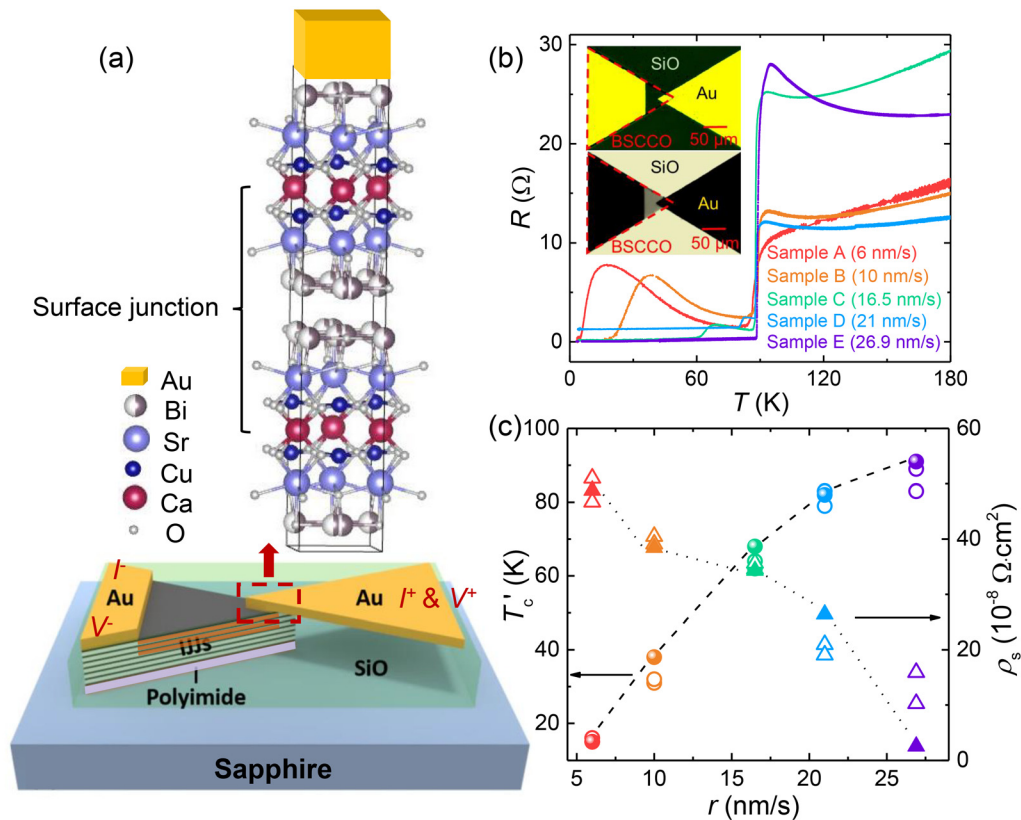


FIG. 1. (a) Schematic structure of SJJ's. (b) and (c) Comparison of samples prepared with different *in situ* gold evaporation rates r . With different colors for different r , graph (b) displays R vs T curves for five selected samples A–E, and graph (c) indicates the junction critical temperatures T_c' and the contact resistances ρ_s for all junctions measured (three devices for each r). In (b), the r values are indicated next to the sample name, and in (c), samples A–E are indicated by solid symbols. Inset in (b) shows typical optical images of the SJJ's. The top one is a positive light image, and the bottom one is a backlight image.

allowing for the possibility to observe a few c -axis junctions in the contacting area between the right gold film and the triangular BSCCO crystal. The SJJ's are located in the diamond-shaped region on the uppermost BSCCO layer with lateral size a (side length) indicated by the dashed rectangle. The junction areas $S = \sqrt{3}/2a^2$ are in the range of 20–450 μm^2 .

Figure 1(b) shows resistance R vs temperature T curves for five selected samples A–E. In the R vs T measurements, the bias current is set to 10 μA . The samples are prepared with the same fabrication conditions except for the evaporation rate of the gold electrodes r during the *in situ* evaporation process. For temperatures above $T_c \approx 91$ K, the measurement picks up both in-plane and out-of-plane components of the resistance of the BSCCO crystal, while at lower temperatures, one observes the resistance of the SJJ's plus the contact resistance between the SJJ's and the Au layer. Below the transition temperature T_c' of the SJJ's, only the contact resistance remains. The T_c' values of samples A–D are 15, 38, 68, and 82 K. For sample E, there is only one superconducting transition temperature of 91 K in the whole temperature range, which means that the SJJ has the same critical temperature as the bulk. On the left axis of Fig. 1(c), we show the T_c' values for all samples measured as a function of r . T_c' increases monotonically with the increase in r . We also find that the product ρ_s of the contact

resistance and the junction area systematically decreases with the increase in r as shown on the right axis of Fig. 1(c). In particular, ρ_s of sample E is about $2.5 \times 10^{-8} \Omega \text{ cm}^2$, which is close to the optimal value obtained for samples cleaved at liquid nitrogen temperature.³³

We next turn to detailed characterizations of the SJJ's at different deposition rates. Figure 2(a) displays an I – V curve (IVC) of severely degraded sample B at 6.6 K. Clearly, the IVC is strongly hysteretic with a critical current I_c of 0.42 mA. The return current I_r is about 90 μA , and the voltage jump $V_c = I_c R$ is around 2.65 mV, which is ten times smaller than that of inner IJJ's observed in other samples. The low value of V_c is mainly due to the reduced critical current density j_c of about 94 A/cm², which is an order of magnitude lower than the j_c of the inner IJJ's. There are no sign of further voltage jumps up to at least 5 mA, making this junction seemingly well separated from the inner IJJ's, without the appearance of additional degraded junctions.

The temperature dependence of I_c and I_r is displayed by solid and open circles in Fig. 2(b) (left axis), respectively. While I_r is almost temperature independent below 16 K, I_c starts to decrease strongly for temperatures above 16 K. Furthermore, for temperatures above 23 K, there is a long tail extending up to about 38 K. This indicates that there is a substantial T_c distribution in the uppermost CuO₂ layer and, consequently, sample B is not very homogeneous at least for temperatures above 16 K.

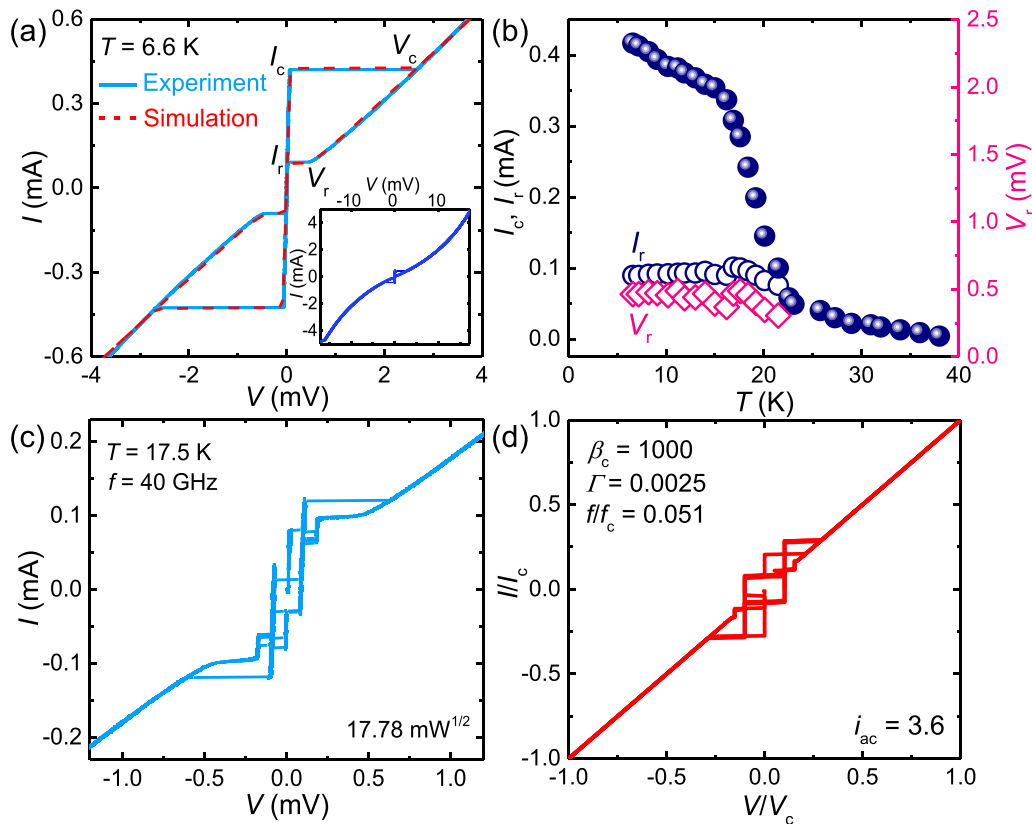


FIG. 2. Data for sample B: (a) IVC measured at 6.6 K. The dashed line indicates simulated IVC based on the RCSJ model. A contact resistance of 0.18 Ω has been added to the simulated curve for better comparison to the data. Inset in (a) shows the 6.6 K IVC on the expanded scale. (b) Critical current I_c (solid circles) and return current I_r (open circles) vs temperature (left axis). Right axis displays by open diamonds; the return voltage V_r vs temperature. (c) IVC in the presence of a 40 GHz microwave field at $T = 17.5$ K. (d) RCSJ simulation corresponding to graph (c).

Next, we derive some average junction properties at 6.6 K. The estimated Josephson length of the SJs is below $0.82 \mu\text{m}$, so this SJJ with a side length a of $22.5 \mu\text{m}$ is clearly in the large junction limit. We further find a value of roughly 26 GHz for the Josephson plasma frequency $f_{\text{pl}} = (j_c s / 2\pi\Phi_0 \epsilon_0)^{1/2}$, where $s = 1.5 \text{ nm}$ is the junction thickness, ϵ_0 is the electric permeability, and $\epsilon \approx 18$ is the dielectric constant.¹⁴

Despite the fact that sample B cannot be considered as a short junction, we do not see signatures of internal modes in the IVCs like the appearance of resonant steps or fluxon-induced features. The IVC can be described well within the resistively and capacitively shunted junction (RCSJ) model as shown in Fig. 2(a) by a dashed line, which considers a short junction without internal degrees of freedom. In the simulation, we have used a McCumber parameter $\beta_c = 2\pi I_0 R^2 C / \Phi_0 = 40$, where C is the junction capacitance and I_0 is the critical current in the absence of thermal fluctuations, and we use the noise parameter $\Gamma = 2\pi k_B T / (I_0 \Phi_0) = 6.6 \times 10^{-4}$.

Generally, within the RCSJ model for $\beta_c \gg 1$, the switch-back from the resistive state to the zero-voltage state occurs at voltage V_r , where the Josephson frequency $f_r = V_r / \Phi_0$ is only slightly larger than f_{pl} . In Fig. 2(b), the temperature dependence of V_r is plotted by open squares. Over a wide temperature range, V_r is close to 0.46 mV and corresponds to $f_r \approx 220 \text{ GHz}$, which is an order of magnitude larger than the value of f_{pl} estimated for $T = 6.6 \text{ K}$. For higher temperatures, the discrepancy is even larger. We think that the SJJ may not behave as an isolated junction but is still strongly coupled to the IJJs stack. Then, the I_r and V_r might be determined by *ac* loss to excitation in the bulk crystal, for example, related to Josephson plasma oscillations in the stack.

More insight can be obtained from the junction behavior under microwave irradiation with frequencies around 30–40 GHz. For an isolated and RCSJ-like junction with a plasma frequency of 200 GHz, one would expect a chaotic behavior without pronounced stable microwave-induced Shapiro steps.³⁴ If the junction behaves RCSJ-like but the I_r is caused by coupling to some collective mode in the inner IJJs, one could expect stable Shapiro steps.

Figure 2(c) shows the IVC for a bath temperature of $T = 17.5 \text{ K}$ and an under microwave irradiation of $f = 40 \text{ GHz}$. At this temperature, the junction characteristic voltage V_c is 1.65 mV, and the characteristic frequency $f_c = V_c / \Phi_0$ is around 780 GHz. Well-behaved Shapiro steps can be observed. The corresponding simulation is shown in Fig. 2(d). Thermal noise is taken into account with noise parameter $\Gamma = 0.0025$. The simulation is at least in qualitative agreement with the experimental observation. For β_c , we use a value of 1000, corresponding to a f_{pl} of 24 GHz. This value is almost the same as the value of 23 GHz estimated from the measured critical current. We further note that in experiment, no stable Shapiro steps appeared for microwave frequencies of 32 GHz or lower.

Overall, the microwave data give good evidence, and sample B is indeed a degraded intrinsic junction, which cannot be considered as isolated but is still electromagnetically coupled to the inner junctions.

Figure 3 displays data for slightly degraded sample C. At 3 K, the SJJ has a critical current I_c of 0.37 mA (a current density j_c of 188 A/cm^2), and the Josephson frequency f_r is about 750 GHz, which is much higher than the Josephson plasma frequency f_{pl} estimated from the j_c like for sample B. The inner IJJs for sample C also have I_c near 3 mA, allowing for a direct comparison of the temperature evolution of the I_c of the SJJ and the inner IJJs, as shown in Fig. 3(b). Evidently, the SJJ is

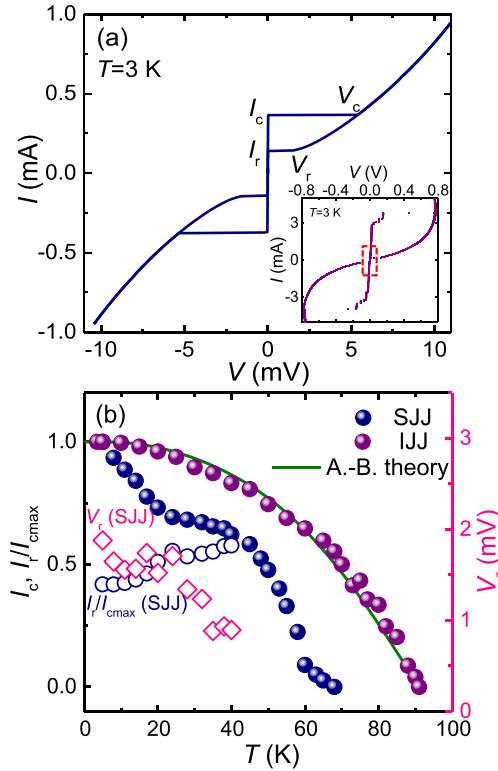


FIG. 3. Data for sample C: (a) IVC measured at 3 K. Inset: IVC on the expanded scale. (b) Temperature dependence of the normalized critical current I_c of the SJJ and the inner IJJs. The solid line represents the temperature dependence of I_c according to the Ambegaokar-Baratoff theory. The return current I_r and return voltage V_r of SJJs are indicated by open circles (left axis) and open diamonds (right axis), respectively.

inhomogeneous, exhibiting two distinct drops in I_c ending near 20 and 60 K, respectively. In contrast, the I_c of the inner IJJs evolves smoothly as a function of temperature, which is close to the dependence expected from the Ambegaokar-Baratoff relation for a superconductor-insulator-superconductor (SIS) tunnel junction.³⁵ For the SJJ, the return current I_r increases with increasing temperature with a somewhat increased slope around 20 K. The return voltage V_r is about constant below 20 K with values of 1.6–1.9 mV ($f_r \approx 770$ –920 GHz). V_r and, correspondingly, f_r decrease for higher temperatures, reaching a value of 0.9 mV (430 GHz) at 40 K, i.e., about half of the respective low temperature values. At this temperature, the I_c of the SJJ is about 60% of its 3 K value, and the I_c of the IJJs has decreased to 80% of the 3 K values. The fact that we do see a temperature dependence of V_r and f_r shows that the return to the zero-voltage state cannot be associated with geometric resonance in either the SJJ or the IJJs, or simply be associated with the excitation of (homogeneous) Josephson plasma oscillations in the IJJs. We will address the issue further at the end.

Finally, Fig. 4 shows data for non-degraded sample E. Figure 4(a) displays the IVC measured at 3 K. A total of eight resistive branches can be identified, representing eight Josephson junctions in series. The I_c of the SJJ is 1.31 mA, which is close to the critical current $I_c = 1.71 \text{ mA}$ of the inner IJJs. Moreover, the voltage jump of the SJJ is up to $V_c = 24.25 \text{ mV}$, almost as high as the characteristic voltage $V_c = 26.1 \text{ mV}$

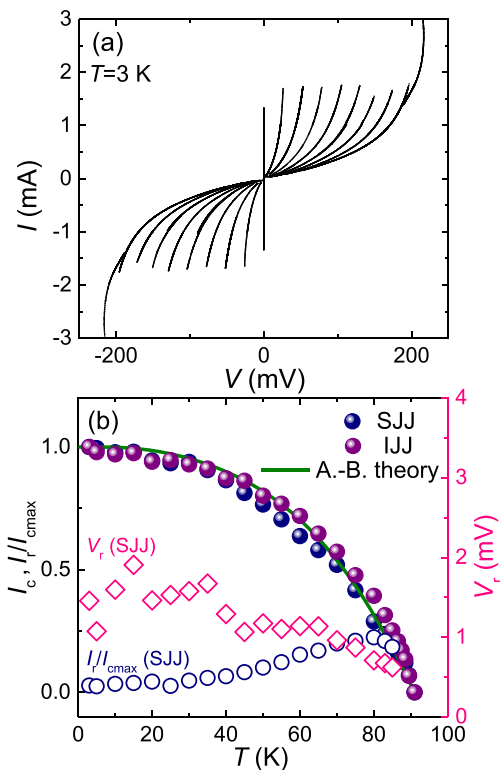


FIG. 4. (a) Data for sample E: IVC measured at 3 K. (b) Temperature dependence of the I_c for the SJJ and the inner IJJs. The solid line represents the I_c vs T according to the A.-B. theory. The I_r and V_r of SJJ are also indicated by open circles (left axis) and open diamonds (right axis), respectively.

of the inner IJJs. The temperature dependence of the I_c of the SJJ and the inner IJJs are displayed in Fig. 4(b). All critical currents follow basically the same temperature dependence and are in good agreement with the A.-B. relation. For the SJJ, we also display the return current I_r and the return voltage V_r . The return frequency f_r is about 1.3 THz and, thus, continues the trend that f_r increases with increasing T_c' and j_c . The I_r of the SJJ smoothly increases with temperature while the V_r is noisy but shows a trend to decrease from 1.7 to 0.6 mV, perhaps in a step-like fashion.

For samples C–E, no microwave-induced Shapiro steps are detectable up to 40 GHz, which indicates that the Josephson plasma frequency is too close to or even higher than the irradiation frequency. Shapiro steps are likely to appear at much higher frequencies, which are out of the present experiments.

This leaves the question about the origin of the switch-back from the resistive to the zero-voltage state. For sample B, the switch-back occurs at a roughly temperature-independent voltage below 20 K. For samples C and E, a clear temperature dependence is observed. In all cases, the return frequency is much higher than the estimated values for both the SJJ and all of the IJJs, ruling out that spatially homogeneous plasma oscillations are excited by the Josephson currents oscillating in the SJJ. For samples C and E, there is some indication that the reduction in f_r with increasing temperature occurs in a step-like manner. This could mean that at different temperatures, the SJJs couple to

different geometrical modes (individual or collective) oscillating in the IJJs. The coupling of the Josephson currents to cavity resonances has been intensively discussed in the context of BSCCO-based THz generators.^{36–45} It should be noted that these generator stacks contain a very large number of IJJs (around 1000), are much bigger in size than the samples discussed here, and are often operated under strong temperature gradients. For these stacks, the coupling to cavity modes is essential to produce strong THz radiation, while the switch-back occurring in the subterahertz range is also produced by some resonances reaching a too-high amplitude. We consider it likely that the excitation of cavity modes in the inner IJJs also plays a role for the electromagnetic behavior of the SJJs, independent of their degradation state.

In conclusion, the motivation of this work is to provide a convenient and reproducible technique to tailor the superconductivity of the uppermost BSCCO layer, and this has been achieved by using a high-vacuum *in situ* evaporation technique at room temperature, allowing for deposition of gold electrodes with a controlled deposition rate immediately after cleaving. In more detail, we have studied the electrical properties of the surface-near layers of BSCCO single crystals. The properties of the two outermost superconducting sheets are characterized by the transport characteristics of the SJJ. The SJJs' critical current density as well as their transition temperature depend systematically and reproducibly on the deposition rate of the gold electrode. This feature allows one to tailor the electric properties of the SJJs according to specific applications smoothly, spanning the range from completely degraded to almost non-degraded. Furthermore, the electromagnetic characterization of these junctions reveals that the SJJs may be coupled to excitations in the inner IJJs' stack. Excitations within the SJJs cannot be ruled out completely. The comprehensive investigation on the transport properties and microwave response of the SJJs tells that one needs to take the electromagnetic coupling among layers into account in the applications of high- T_c BSCCO single crystals.

Our *in situ* evaporation technique yields reliable results without the need of complex fabrication steps, paving the way for practical applications of high-temperature superconducting electronic devices.

See the [supplementary material](#) for the detailed fabrication process of the SJJs, the geometrical and some electrical parameters of the selected five samples, estimation of the Josephson length of the SJJs, and the transport properties of samples A and D.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Zihan Wei: Data curation (lead); Writing – original draft (lead). **Dieter Koelle:** Writing – review & editing (supporting). **Reinhold Kleiner:** Formal analysis (equal); Writing – review & editing (equal).

Huabing Wang: Funding acquisition (equal); Writing – review & editing (equal). **Peiheng Wu:** Funding acquisition (supporting). **Hongmei Du:** Software (equal); Writing – review & editing (equal). **Dingding Li:** Data curation (supporting); Software (supporting); Writing – review & editing (supporting). **Meiping Jiang:** Data curation (supporting). **Ping Zhang:** Writing – review & editing (supporting). **Shixian Chen:** Writing – review & editing (equal). **Yang-Yang Lyu:** Writing – review & editing (supporting). **Hancong Sun:** Writing – review & editing (equal). **Yong-Lei Wang:** Writing – review & editing (supporting).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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