# **Intrinsic Josephson junctions in misaligned**  $Tl_2Ba_2CaCu_2O_{8-r}$  **thin films with different tilt angles**

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Tl<sub>2</sub>Ba<sub>2</sub>CaCu<sub>2</sub>O<sub>8-*x*</sub> films on vicinal LaAlO<sub>3</sub> having different misorientation angles (10°, 15°, 20°, 25°) grow with their  $CuO<sub>2</sub>$  planes tilted with respect to the substrate surface. We found intrinsic Josephson behavior in microbridges patterned out of these films for misorientation angles equal to or larger than 15°. Here we report on the fabrication process, structural, and electrical properties. Structural analyses include secondary electron microscopy images and x-ray analysis of the crystallographic structure. We also present the current voltage characteristics, which exhibit a different behavior for the 15° films than for 20° and 25° films: 15° films show enhanced critical currents, while 25° films show giant collective switching events, indicating a very small spread of critical currents. We discuss these effects with the possibility of shunts in the case of the 15° films and with the existence of collective modes in the case of the 25° films. In the latter case, we also discuss the possibility of synchronization.

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# **I. INTRODUCTION**

High temperature superconductors (HTSC)-especially the thallium- and bismuth-based cuprates—are characterized by a number of outstanding properties. Their layered crystal structure with very thin superconducting copper oxide planes leads to a strong anisotropic behavior and a *c*-axis current transport, which is dominated by intrinsic Josephson effects.<sup>1</sup> The large gap voltages of cuprates are the produce the high characteristic frequencies of their Josephson junctions. Appliances which use these intrinsic Josephson effects are promising candidates for local oscillators and other devices, which operate in the high GHz or THz range. $2$ 

The fundamentals of this technology have been established in the last decade. Initial measurements were made on  $Bi_2Sr_2Ca_3Cu_2O_{10}$  single crystals.<sup>1</sup> Some years later, the first measurements on thin films were presented[.3](#page-7-3) Despite a number of successful investigations, these layouts still contain problems related to their normal conducting electrodes, which lead to additional low frequency noise $4$  and charge imbalance effects.<sup>5</sup> This problem has been solved in the last few years by the use of different types of technology. Wang *et al.*[6](#page-7-6) patterned single crystals in a double-sided process, while Kim *et al.*<sup>[7](#page-7-7)</sup> used a focused ion beam to pattern mesalike structures out of thin films. An easier way to pattern intrinsic Josephson junctions was developed which involves depositing thin films on vicinal cut substrates. $8,9$  $8,9$  On such substrates the films grow with their copper oxide planes tilted with respect to the substrate surface and the intrinsic Josephson effects can be measured in simple bridgelike structures, which have superconducting feed lines and can easily integrated in high frequency schemes.

In addition to solving the technological problems, a theoretical understanding of intrinsic Josephson junctions was also established. Models which involve a magnetic interlayer coupling were developed in the first publications.<sup>10[,11](#page-8-0)</sup> Some years later the idea of electrical interlayer coupling was introduced.<sup>12</sup> Since 1997 a theory of the temperature dependence of the *c*-axis current transport in strong anisotropic HTSC has also been available.<sup>13</sup> This theory takes into account the  $d_{x^2-y^2}$  symmetry of the order parameter of the cuprates. An important problem for microwave applications is the question of phase locking, which is necessary for the emission of a sufficient power of radiation. Previously, we have published a number of papers about this topic especially dealing with phase locking in resonant environments and in the presence of resistive shunts. $14,15$  $14,15$  In this paper we present Tl-2212 films on vicinal cut LaAlO<sub>3</sub>. The results of Tsai *et al.*<sup>[8](#page-7-8)</sup> for very small misorientation angles  $(2^{\circ})$  showed only a reduction of critical currents but no sign of intrinsic Josephson effects, while in 20° films intrinsic Josephson ef-fects could be observed.<sup>16,[17](#page-7-6)</sup> So it was our intention to test films having different misorientation angles with respect to the influence of this angle on the electrical properties, especially on the shape of the current-voltage characteristics and the critical currents. We characterize here the structural and electrical properties on 10°, 15°, 20°, and 25° LaAlO<sub>3</sub>.

The paper is arranged as follows: In Secs. II and III, we describe the preparation of the thin films and their structural properties. In Sec. IV, after some preliminary remarks, we explain the setup for the electrical measurements and present their results. Section V is dedicated to the discussion of the results and Sec. VI is the conclusion.

# **II. FILM PREPARATION**

High temperature superconducting  $Tl_2Ba_2CaCu_2O_{8-r}$  (Tl-2212) thin films were prepared on vicinal lanthanum aluminate substrates (LaAlO<sub>3</sub>) with misorientation angles of  $\Theta$  $=10$ , 15, 20, and 25°. Epitaxial growth of the Tl-2212 thin film on the vicinal substrates creates a misalignment of the copper oxide double planes of the superconductor by the angle  $\Theta$  with respect to the substrate surface. A current passing through a small bridge structure aligned in the  $(\cos \Theta \ 0 \sin \Theta)$  direction will contain a *c*-axis transport current component. In this way, the observation of intrinsic Josephson effects is possible.

Since the extremely high volatility of the thallous oxide

(Tl<sub>2</sub>O) prevents a simple *in situ* deposition technique, we used a two-step process<sup>18</sup> for fabricating Tl-2212 thin films. First, we deposited amorphous thallium-free precursor films of  $Ba_2CaCu_2O_v$ . For this purpose we used a reactive magnetron sputtering technique in special off-axis geometry with a metallic alloy target having a composition close to  $Ba<sub>2</sub>CaCu<sub>2</sub>$ . The off-axis geometry prevents the negatively charged oxygen ions generated in the sputtering discharge from reaching the substrates. The growing films are thus protected against detrimental energetic particle bombardment which would lead to composition changes due to massdependent sputtering yields. The sputtering target was powered in dc mode at 0.9 W/cm2 target power in an argon/ oxygen atmosphere of 0.035 mbar. During deposition the substrates were kept at about 250 °C in order to avoid water contamination and improve thin film adhesion. We deposited films with thicknesses between 100 and 205 nm. The resulting precursor films show a nearly 2:1:2 cation composition with a slight excess of copper, which is desired for a better temporal stability of the precursor films. Precursor films with exactly a 2:1:2 composition are very sensitive to humidity because of the large amount of the chemically very reactive elements barium and calcium. The precursor films are electrically insulating and as smooth as glass. The stoichiometry of the precursor films was controlled by electron stimulated x-ray microanalysis in the energy dispersive mode (EDX) by means of a "JEOL-8800L Superprobe" instrument. In order to reduce the influence of the substrate material on the analyzed volume for measurements of small film thickness, the excitation energy of the scanning electron microscope was decreased from the normally used 15 keV to 12 keV. This shortens the mean free path of electrons in the sample. The same instrument was used for SEM inspection. In the second step, oxy-thallination, thallous oxide  $(Tl_2O)$  was incorporated into the precursor film which induces crystallization to the Tl-2212 phase via a solid-state reaction. For this purpose the Ba<sub>2</sub>CaCu<sub>2</sub>O<sub>y</sub> precursor films were annealed at ca. 860 °C in a  $T_2O+O_2$  atmosphere and subsequently cooled under flowing oxygen. The annealing was performed in a furnace with the precursor film and a Tl-Ba-Ca-Cu-O ceramic pellet enclosed in an Au/Pt-container. The Tl-Ba-Ca-Cu-O ceramic pellet contains  $Tl_2O_3$  which serves as the  $Tl_2O$  source at temperatures above 500 °C via the reaction  $Tl_2O_3^{\text{solid}} = Tl_2O^{\text{gas}} + O_2$ . The crux of the oxy-thallination process is a temperature-time regime, which ensures the correct temperature for the ceramic pellet to release the right amount of  $Tl_2O$  and, at the same time, a substrate temperature, which allows crystallization to the Tl-2212 phase after Tl incorporation into the precursor without the formation of undesired secondary phases. More details on the preparation technology are given elsewhere.<sup>19</sup>

The substrate size of the vicinal  $LaAlO<sub>3</sub>$  was 1 cm<sup>2</sup>. After HTS thin film preparation a 100 nm thick gold layer was sputtered onto the Tl-2212 to protect the HTS film and to ensure good ohmic contacts for the electrical measurements. The gold on top of the region where the vicinal bridges were patterned was removed by argon ion beam etching. The crystallographic orientation, especially of the vicinal tilting of the *c* axis, was examined using x-ray diffraction measurements made with a Philips X Pert diffractometer.

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FIG. 1. Pole figures of (a) a 205 nm thick vicinal Tl-2212 thin film with mixed orientations on  $10^{\circ}$  vicinal LaAlO<sub>3</sub> and (b) a 160 nm thick perfectly oriented vicinal Tl-2212 thin film without  $c$ -axis orientation on  $20^\circ$  vicinal LaAlO<sub>3</sub>.

#### **III. FILM PROPERTIES**

The vicinal substrate should lead to a vicinal HTS thin film orientation. In fact, we can obtain tilted CuO double planes in such films. Unfortunately, the HTS films did not always crystallize in a tilted orientation which corresponded to the vicinal substrates. In the substrate plane the vicinal substrate provides the geometrical information for vicinal film growth due to the miscut angle  $\Theta$  along one direction (the direction of the later patterned bridge), whereas in the orthogonal direction the growing film sees the nondisturbed *a* axis of the substrate. The difficult vicinal crystallization of Tl-2212 is due to the two-step process of the Tl-2212 thin film preparation. The Tl-2212 thin films are not prepared *in situ* (as in the case, e.g., for  $YBa_2Cu_3O_{7-x}$ ), but the amorphous precursor is transformed into the superconducting 2212 phase. Consequently, small variations in annealing conditions such as temperature-time regime, substrate temperature, or  $Tl_2O$  supply, or in precursor film composition may favor the conventional *c*-axis crystallization of the Tl-2212 thin film in some areas instead of vicinal crystallization *c*-axis orientation means that the *c* axis of the Tl-2212 thin film is oriented normal to the substrate surface).

For  $\Theta = 10^{\circ}$  we primarily observed *c* axis rather than vicinal crystallization. The misorientation angle of the substrate is sufficiently low that the kinetics of the epitaxial Tl-2212 nucleation is nearly undisturbed. Figure  $1(a)$  $1(a)$  shows an x-ray pole figure of a 205 nm thick Tl-2212 thin film on 10° vicinal La $AIO_3$ . The four larger pole densities symmetrically arranged around the origin represent the *c*-axis orientation, while the smaller four pole densities below them result from the 10° tilted growth.

For  $\Theta = 15^{\circ}$  and  $20^{\circ}$  we found the desired vicinal orientation in the films, sometimes with small *c*-axis oriented areas. This is consistent with the SEM and x-ray inspections. Figure  $1(b)$  $1(b)$  shows the x-ray diffraction (XRD) pole figure of a perfectly grown 20° vicinal Tl-2212 thin film without *c*-axis oriented parts, whereas Fig. [2](#page-2-0) shows a SEM image of a Tl-2212 thin film on  $20^{\circ}$  LaAlO<sub>3</sub> with mixed orientation.

For  $\Theta = 25^{\circ}$  we clearly obtained vicinal orientation, but in some cases the films show interruptions, because the misorientation angle is too large.

Furthermore, we also observed that the amount and size of the *c*-axis-oriented regions decreases with increasing film

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FIG. 2. SEM image of a Tl-2212 thin film with mixed orientation on 20 $^{\circ}$  vicinal LaAlO<sub>3</sub> with vicinal (V) and *c*-axis (C) oriented areas.

thickness (from 100 to 205 nm). A possible reason for this is that thinner films are able to compensate for the mechanical stress and so crystallize *c*-axis oriented even on a vicinal substrate, whereas thicker films preferentially crystallize in the vicinal orientation.

The critical temperature,  $T_c$  of the vicinal superconducting films was measured using an inductive technique. By monitoring the temperature dependent real part of the ac susceptibility  $\chi'(T)$  we can observe the transition from the normal to the diamagnetic, i.e., superconducting state, and thus obtain the critical temperature. The onset temperature of 98–103 K was independent of the misorientation angle  $\Theta$ , but the transition width increased with increasing misorientation angles  $\Theta$ .

### **IV. ELECTRICAL PROPERTIES**

#### **A. Preliminary remarks**

At finite temperatures the switching of intrinsic Josephson junctions from the superconducting to the quasiparticle branch is a stochastic process. Thermal noise and electrical perturbations lead to the destabilization of the superconducting state near the critical current  $I_{\rm C}$  and the junction switches sooner or later from the superconducting to the resistive state depending on the amount of noise. During repeated recordings of the current-voltage dependence, we obtain not a single value for the switching currents  $I_{S\uparrow}$  and  $I_{S\downarrow}$ , but a switching current distribution  $P(I_{S\uparrow})$  and  $P(I_{S\downarrow})$ . Here we call  $I_{S\uparrow}$  the bias current where switching from the superconducting to the resistive state occurs and  $I_{S\parallel}$  the bias current where switching from the resistive to the superconducting state occurs. The switching current distribution may be narrow for a single Josephson junction. However, for an array of many junctions, as in our case, we have to take into account the mutual interaction of the Josephson junctions, which can act as electrical perturbation and lead to a broadening of the switching current distribution. For the evaluation of this distribution we use at least 50 complete bidirectionally recorded current-voltage measurements. In order to appraise the critical currents  $I_{\rm C}$  of our samples we give both the mean values as well as the standard deviation of the switching current distribution in this paper. In individual cases we also give the maximum switching current. The return current  $I_R$  is handled in the same manner.

Our thin films have a large surface roughness. In a previous publication<sup>16</sup> we presented the results of a transmission electron microscopy and atomic force microscopy study. These results indicate that our Tl-2212 microbridges can be modeled as a serial array of serial subarrays of intrinsic Josephson junctions. The length scale of the array is the length of the microbridge itself, which is 2  $\mu$ m. The length scale of the subarrays is in the range of some hundred nanometers. The parameter spread film thickness, critical current, barrier resistance, etc.) between the subarrays may be large due to geometrical variations of the cross section, but the parameter spread in one subarray can be accounted as small. Thus, the rough films permit electrical measurements with a good reproducibility. We acquire area-specific parameters such as critical current density  $j<sub>C</sub>$  or the normal resistivity  $\rho<sub>N</sub>$  in the reproducible lower part of the current-voltage dependence, i.e., for the junctions with the smallest critical currents. These junctions are in the narrowest part of the microbridge, and therefore, the values for  $j<sub>C</sub>$  will be minimum values whereas the values of  $\rho_N$  will be maximum values. One difficulty in determining the specific parameters, which mostly depend on thickness, is the measurement of the local geometrical parameters such as thickness or width.

#### **B. Measurement setup**

The microbridge structures with lengths 1 of 2  $\mu$ m (with the exception of the 20 $\degree$  sample where bridges were 10  $\mu$ m long), and widths  $w$  of 2  $\mu$ m for all samples were patterned by photolithography and  $Ar^+$  ion beam milling from a Tl-2212 film of 205 nm  $(120 \text{ nm}$  for  $20^{\circ}$  sample) thickness. The bias feed lines were patterned in the same way.

The bridges contain serial arrays of Josephson junctions with circa 230 and 440 junctions for 15 and 25°, respectively, accordingly to the equation

$$
N = (l \sin \Theta - h \cos \Theta)/(s + t), \tag{1}
$$

where N is the number of junctions, *l* and *h* are length and height of the bridge, *s* and *t* are the thickness of the interplanar separation layer and the copper oxide plane, and  $\Theta$  is the tilt angle of the copper oxide planes<sup>9</sup> (see Fig. [3](#page-3-0)). The  $20^{\circ}$ samples contain circa 2000 junctions, and are described in more detail in Ref. [16.](#page-8-5)

The electrical measurements were made using a two-point geometry (four-point geometry for the 20° sample). The large advantage of our films is the superconducting bias feed lines. Due to the large contact pads far away from the microbridge, the contact resistance between the gold and Tl-2212 is negligible. Quasiparticle injection into the microbridge, which leads to additional fluctuations in mesa type intrinsic Josephson junctions<sup>4</sup> is prevented. The wires were shielded

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FIG. 3. Sketch of a vicinal bridge with length *l*, width *w*, HTS thin film thickness  $h$ , and tilting angle  $\Theta$ . The hatched area in the bridge denotes the tilted copper oxide double planes. The large arrow denotes the current flowing through the vicinal bridge.

by grounded covers. The electrical supply and the measurements were made by a low noise source and measurement unit from Keithley. The electrical characterization was performed both current-driven and voltage-driven. This enables a better imaging of the spread of the switching currents, even in the presence of collective switching events due to the very small spread of the critical current. In order to account for the statistical behavior of the switching into the resistive state we measured each I-V dependence 50 times. To record the hysteretic behavior all dependencies were measured in a bidirectional way. The measurements were performed either in liquid helium or in a continuous flow cryostat. The sample where shielded by a Mu-metal shield to reduce magnetically disturbances by the environment.

#### **C. Measurement results**

#### *1. Critical temperature*

We performed the temperature dependent measurement of the resistance on the same  $2 \times 2 \mu m$  bridges which were used for the current-voltage measurements. On the 15° and 25° films we used a probe current of 100  $\mu$ A and 1  $\mu$ A, respectively. In the temperature range from 300 K to about 120 K, the bridges show a metallic behavior. Below 120 K the resistance decreases rapidly. The critical temperatures are 114 K  $(T_{\rm C}^{90\%})$ , 99 K  $(T_{\rm C}^{10\%})$ , and 90 K  $(T_{\rm C0})$  for the 15° sample and 112 K  $(T_{\rm C}^{90\%})$ , 97 K  $(T_{\rm C}^{10\%})$ , and 89 K  $(T_{\rm C0})$  for the 25° sample. The values  $T_{\rm C}^{90\%}$ ,  $T_{\rm C}^{10\%}$  are related to the 10  $\mu$ m feed lines and broader structures, while the value  $T_{C0}$ originates from the 2  $\mu$ m bridge structure. The  $T_{C0}$  of the 20° sample is 90 K.

#### *2. Current-voltage dependency and critical currents*

The current-voltage dependency of the microbridges show branch structures (Fig. [4](#page-3-1)), which are typical for serial arrays of intrinsic Josephson junctions. This structure of multiple branches is caused by a step-by-step switching of intrinsic Josephson junctions during the current sweep due to the spread of critical current or even for no spread due to nonequilibrium effects such as charge imbalance and collective charging of the superconducting layers.<sup>20,[23](#page-8-9)</sup> The branches in our measurements are mostly caused by the spread of critical current. However, we found some peculiarities related to the different misorientation angles.

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FIG. 4. Accumulated (20 single bidirectional measurements) current-voltage dependencies of  $2 \times 2$   $\mu$ m microbridges of Tl-2212 films on vicinal LaAlO<sub>3</sub> with misorientation angles of  $15^{\circ}$ (a) and 25° (b). The 15° films show distinctly enhanced critical currents.

The 15° microbridges show small collective switching operations mostly between 10 to 20 mV. The backward tracing of the current-voltage dependency show that these collective events are due to two to six single junctions, which return into the superconducting state both individually as well as in collectively. In current-driven measurements these collective switching operations appear as zones without branches in the hysteretic regions. However, voltage-driven measurements show that branches exist in this region, which are not stable in the current-driven case. The branch which is associated with the lowest  $I_{S\perp}$  value shows the largest return interval in the voltage range. All other return intervals in the lowest hysteretic region are smaller. This is a typical behavior for a serial array of Josephson junctions with a shunt.<sup>15</sup> The mean values of the  $I_{S\uparrow}$  distributions for different bridges range from 150  $\mu$ A to 300  $\mu$ A. A typical value is 200  $\mu$ A. The standard deviation of the  $I_{S<sup>†</sup>}$  distribution is in the range of 10  $\mu$ A. In one atypical case we measured a switching current distribution with a mean  $I_{S\uparrow}$  value of 820  $\mu$ A and a standard deviation of 30  $\mu$ A. The  $R_N$  values range from 10 to 100  $\Omega$ . The  $I_C R_N$  products of 10 to 20 mV per junction are in good agreement with literature values.<sup>17</sup> However, we measured  $I_{\rm C}R_{\rm N}$  products of smaller than 6 mV as well. This is also an

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FIG. 5. Accumulated (10 single bidirectional measurements) current-voltage dependencies of  $2 \times 2$   $\mu$ m microbridges of Tl-2212 films on vicinal LaAlO<sub>3</sub> with a misorientation angle of  $25^\circ$ . Measurements are current driven in (a) and voltage driven in (b). Due to the small spread of the critical currents and collective modes the branches which are measured in the voltage-driven case are unstable in the current driven case. This leads to giant collective switching events over 2.5 V (100-150 Josephson junctions).

indication of the presence of shunts. An important parameter of the resistively and capacitively shunted model of Jose-phon junctions (RCSJ-model)<sup>[21](#page-8-10)</sup> is the McCumber parameter  $\beta_c$  which determines the hysteretic behavior and which can be estimated by the ratio  $I_{S\uparrow}/I_{S\downarrow}$ . We found it to range from two to four, but for some junctions it was up to 20. The standard deviation of the  $I_{S\perp}$  distribution was smaller than that of the  $I_{S\uparrow}$  distribution.

For the 20° microbridge we measured small collective switching events. However, there were no signs of shunts. The mean value of the smallest  $I_{S\uparrow}$  is 18  $\mu$ A, with standard deviation of 1.5  $\mu$ A. The  $R_N$  values range from 1 to 2 k $\Omega$ . The  $I_C R_N$  products are 15–20 mV and the values of  $\beta_C$  are approximately 20.

For the 25° microbridges we observed small collective switching events  $(2-5$  junctions) as well as individual switching events. For a few of the bridges we found even giant collective switching operations over a voltage range of 1 to 5 V, which represents a simultaneous or nearly simulta-

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FIG. 6. Time dependent measurement of the giant collective switching events reveals their successive character. It comprises a number of smaller collective switching operations.

neous switching of 50 to 300 junctions from the superconducting state into the resistive state [Fig.  $5(a)$  $5(a)$ ]. Voltagedriven measurements [Fig.  $5(b)$  $5(b)$ ] show the existence of branches inside the giant hysteresis. These branches are not stable in current-driven measurements. With current-driven time-resolved measurements (Fig.  $6$ ) we could identify the giant collective switching events as a fast succession of smaller collective events (2-50 junctions). Here "fast" is used comparable to the time resolution of current-driven measurements of the current-voltage dependence. The mean values of  $I_{S\uparrow}$  are typically in the range around 30  $\mu$ A, with a standard deviation of 1.5  $\mu$ A. The  $R_N$  values range from 500 to 1000  $\Omega$ . The  $I_C R_N$  products are 10–20 mV and values of  $\beta_c$  range between two and ten.

Intrinsic Josephson junctions were found in films with all misorientation angles ( $15^\circ$ ,  $20^\circ$ , and  $25^\circ$ ). The  $I_C R_N$  products are in the range from 10 to 20 mV. Smaller values from the 15° films can be explained by shunts, whose nature will be discussed in Sec. V A. The minimum values of the critical current densities  $j_{\text{C}}$  can be estimated as 50 kA/cm<sup>2</sup>, 8 kA/cm<sup>2</sup>, and 8 kA/cm<sup>2</sup> for 15°, 20°, and 25° films, respectively. The maximum values of normal resistivities are 0.02  $\Omega$  m, 0.2  $\Omega$  m, and 0.3  $\Omega$  m for 15°, 20°, and 25° films, respectively. In summary, we note that 15° films tend to have higher critical current densities and smaller normal resistivities, whereas 20° and 25° films show a similar behavior with lower critical current densities and higher normal resistivities.

We did not completely characterize all 48 microbridges per chip. However, we can estimate that for the 15° films approximately 25% of the bridges show intrinsic Josephson junction behavior whereas for the 25° films approximately 50% of the bridges show intrinsic Josephson junction behavior. These estimations are in agreement with inspections done by light microscopy which was used to control the existence of the vicinal oriented areas inside the bridge region.

#### *3. Temperature behavior*

Figure [7](#page-5-0) shows histograms for the switching events with the smallest  $I_{S\uparrow}$  for a 15° sample at different temperatures. In

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FIG. 7. Temperature dependence of the switching current distribution of a  $2 \times 2 \mu m$  microbridge of Tl-2212 film on vicinal LaAlO<sub>3</sub> with a misorientation angle of  $15^\circ$ . (For further description see text.)

addition to the expected reduction of the switching currents with increasing temperature, we also see a narrowing of the switching current distribution for temperatures above 21 K. For a better understanding of this effect, a more detailed analysis of these switching events is necessary in order to observe at which point the system switches to which particular quasiparticle branch. The results for 4 K and 31 K are shown in Fig.  $8.$  For  $4K$  it can be seen that the broader switching current distribution can be split into a number of smaller distributions. We obtain these smaller subdistributions if we conclude switching operations which switch into quasiparticle branches of the same range. The 9 K and 21 K measurements lead to the same conclusions, and the widths of the subdistributions are comparable to the widths of the distributions at higher temperatures. It is remarkable that switching events which lead to higher quasiparticle branches occur at higher switching currents. This means that the number of junctions which participate in a collective switching event increases with the switching current. This has consequences for the consideration of the temperature dependence of the critical currents which will be discussed in Sec. V. For temperatures above 21 K a partitioning of the distributions is not possible. All switching events lead into the same region of the quasiparticle branches and the distributions in Fig. [4](#page-3-1) become narrower.

#### **V. DISCUSSION**

# **A. Nature of shunts and the origin of the high critical currents of the 15° films**

Two types of resistive shunts are possible: external and internal shunts. The existence of an external shunt can be explained by an incomplete removal of the gold layer. The angular dependence of the sputter plasma during the film deposition is different from that of the  $Ar<sup>+</sup>$  ion beam during the etching process. This may lead to shadowing effects, which would leave small areas on the sample with incomplete gold removal. Since no gold residues could be found by light microscopy and since the resistivity of the bridges at room temperature did not indicate the presence of resistant

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FIG. 8. Accumulated (50 single bidirectional measurements) current-voltage dependencies of  $2 \times 2 \mu m$  microbridges of a Tl-2212 film on vicinal LaAlO<sub>3</sub> with a misorientation angle of  $15^{\circ}$ (gray). The black dots indicate the voltage points on a quasiparticle branch to which the system switches directly from the superconducting branch in a single measurement. In the cases where more junctions switch together the switching current is increased. Consequently the broader switching current distributions at small temperatures are show to be a combination of smaller distributions.

shunts, these areas must be very small. If there are such gold residues, they will act as resistive shunts for a small group of Josephson junctions, which is not necessarily a disadvantage. We showed in previous publications<sup>15[,22](#page-8-11)</sup> that such resistive shunts are able to promote phase locking if their resistances are in the range of the normal resistance of the Josephson junctions.

A second possibility is the existence of internal shunts. Such internal shunts may be provided by several kinds of disturbed areas. For instance, as described in Sec. III, even in films with low misorientation angle, we found areas that show the typical morphology of *c*-axis films, i.e., with the *c*-axis perpendicular to the surface normal. This can be observed by light microscopy. We cannot exclude the possibility that besides such microscopic *c*-axis fractions, nanoscopic *c*-axis fractions might exist as well, which are not detectable in light or secondary electron microscopy. In the same manner, we can discuss other disturbed areas such as interface layers and grain boundaries.<sup>16</sup> If such areas are located alongside or on top of the bridge, they act as resistive or even as superconducting shunts and could explain the branch narrowing. The higher critical current densities of the 15° films can also be explained with such nanoscopic shunts.

#### **B. Collective switching processes**

The analysis of the switching current distributions shows that an evaluation of the temperature behavior of the intrinsic Josephson junction arrays is only possible after considering the nature of the switching events.

For *T*=0 all Josephson junctions are in the superconducting state and their phase difference rests in a local minimum of the washboard potential. $^{21}$  In the case of intrinsic Josephson junctions, the superconducting  $CuO<sub>2</sub>$  planes are thin in comparison to the charge screening length. If the critical current is exceeded for one junction, its phase difference starts to rotate. This causes charging effects in adjacent junctions<sup>12[,20](#page-8-8)</sup> and the phase differences of these junctions in turn begin to oscillate. This oscillation leads to a coupling of the junctions, which can no longer be considered to be independent from each other. Different quasiparticle branches are associated with different patterns of rotating and oscillating junctions.<sup>23</sup> For small finite temperatures, the superconducting state shows thermal activated oscillations of the phase difference.

The collective switching events can be explained as follows: An initial junction starts to rotate, which causes oscillating phase differences in the neighboring junctions. The strength of these oscillations increases the closer these neighbors are to the rotating junction. These neighbors have only a slightly higher critical current than the rotating junction. This assumption is consistent with the model of the microbridge as an array of subarrays as described above. We assume that the spread of the critical currents between the junctions in a subarray is very small. This again is consistent with the results of the voltage-driven measurements [see Fig.  $5(b)$  $5(b)$ ]. If the neighbors of the rotating junction have only a slightly higher critical current, their washboard potential is so far tilted that even a small perturbation, such as an induced additional oscillation, is sufficient to raise these neighboring junctions also into the rotating state.<sup>24</sup> The  $\beta_c$  values from two to ten are sufficient to keep the return current small enough for the stabilization of the rotating state. The process of a successive switching of the neighboring junctions will continue until a junction with a much higher critical current is reached. The smaller (that means: not giant) collective switching events, which are over voltage ranges, and which are associated with 2–50 junctions, support this explanation.

Another indication for the existence of a coupling is the fact that, for low temperatures, and for the first transition from the superconducting state into the quasiparticle branch region, the switching current scales with the number of junctions that switch together in a collective mode. The higher switching currents for larger collectives can be explained by an occasionally acting mechanism that reduces the switching current and inhibits the coupling in the collective. Appropriate mechanisms for this are external perturbations or different kinds of fluxon modes. The latter case is possible since the Josephson pentration depth  $\lambda_J$  can be estimated to ap-

proximately 0.1  $\mu$ m (Ref. [25](#page-8-13)) which is smaller than our sample dimensions and magnetic field from external disturbances or frozen flux during the cooling process may cause pancake vortices at least in the superconducting feed lines in the vicinity of the microbridge. In order to clarify the correct mechanism, measurements in different magnetic fields need to be done. Mros *et al.*[26](#page-8-14) as well as Warburton *et al.*[27](#page-8-15) show bimodal switching current distributions. Mros *et al.* measured mesa stacks in an external magnetic field and explained the distribution with phase locking in the case of two junctions showing collective events. Warburton *et al.* picked up these arguments and explained the switching current distributions of microbridges on vicinal cut substrates without an external magnetic field in terms of phase locking as well. Both authors support their explanation with a thermal activation model. This is a conclusive argumentation, but a stringent proof for phase locking should be supported by the verification of the power dependence on the number of active junctions. $28,29$  $28,29$  Phase locking is very likely to occur if we consider the induced oscillations and the small spread of critical currents.<sup>30</sup>

#### **C. Temperature dependence of the critical current**

In accordance with the assumptions made above, collective switching events of a large number of Josephson junctions at high switching currents are expected in systems with small perturbations only. For higher temperatures, the first transition from the superconducting state into the quasiparticle branches is only caused by transitions of single junctions or small numbers of junctions. This indicates that thermal induced effects are sufficient to reduce the switching current as well as to inhibit the collective switching events. A direct influence of the temperature as described by Franz *et al.*[31](#page-8-19) can be excluded because the Josephson energy  $E_J = \hbar I_C/2e$  of our junctions is at least two orders of magnitude higher than the thermal energy  $E_T = k_B T$ . However, it is conceivable that thermal activated fluxon motion constitutes a mechanism which is able to reduce the switching current as well as to inhibit collective switching events. This model will be tested in further experiments, especially with reference to the coupling of the junctions and to the possibility of synchronization. In the  $I_C$ -*T* diagram (Fig. [9](#page-7-11)) we see a switching current reduction for temperatures of 31 K and higher. The drawn error bars (black bars) give the standard deviation of the whole switching current distribution. For temperatures of 31 K and above these standard deviations are narrowed due to the absence of the collective switching events at higher current values. Without mechanisms which prevent collective events, the switching currents should also reach higher values at temperatures above 31 K. In order to demonstrate the magnitude of this effect we include error bars (gray bars) in Fig. [9](#page-7-11) which we estimated on the basis of the standard deviations of the switching current distributions for lower temperatures. The critical currents are located on the top of these bars. To compare these with theoretical predictions we drew two theoretical lines in Fig. [9](#page-7-11) as well. The first shows the  $I_{C}$ -*T* dependence of Ambegaokar and Baratoff<sup>32</sup> which is often used in the theoretical predictions for superconductor-

<span id="page-7-11"></span>

FIG. 9. Temperature dependence of switching currents of  $2 \times 2$   $\mu$ m microbridges of Tl-2212 film on vicinal LaAlO<sub>3</sub> with a misorientation angle of 15°. The lines show the theoretical predictions for superconductor-insulator-superconductor junctions [Ambegaokar and Baratoff (Ref. [31](#page-8-19)), solid line] and for *c*-axis transport in highly anisotropic superconductors Tanaka and Kashi-waya (Ref. [13](#page-8-2)), dotted line]. The inset shows the shifting of the switching current distributions (see text), which lead to the reduction of the switching current mean values for higher temperatures.

insulator-superconductor (SIS) junctions. However, this theory is restricted to *s*-wave superconductors. A more realistic theory for the *c*-axis transport in strongly anisotropic high temperature superconductors can be found in the paper of Tanaka *et al.*<sup>[13](#page-8-2)</sup> The  $I_{C}$ -*T* dependence of this reference is based on a superconductor-insulator-superconductor model and takes into account the  $d_{x^2-y^2}$  wave symmetry of the su-perconductor (see also Fig. [9](#page-7-11)). Remaining deviations can be explained by the consideration of disturbed regions (i.e., low angle grain boundaries, phases with other stoichiometry, etc.). Depending on their position inside the microbridge, such regions are able to influence the transparency of the barrier between the  $CuO<sub>2</sub>$  planes.

# **VI. CONCLUSION**

We fabricated thin films of Tl-2212 on vicinal cut  $LaAlO<sub>3</sub>$ with misorientation angles of 10°, 15°, 20°, and 25°. While the 10° films show only few vicinal growth areas, in films with higher misorientation angles we found a high number of regions with tilted copper oxide planes. We patterned microbridges out of the films with misorientation angles of 15°, 20°, and 25° and found intrinsic Josephson behavior in all cases. The 15° films showed enhanced critical current densities and diminished resistivities in comparison to films with 20° or 25°. We found clear signs of shunting in some of the microbridges patterned in 15° films, which can be explained by internal and external shunts. The microbridges from 20° and 25° films showed similar behavior, i.e., the critical current densities and the resistivity are in the same range. A particularity of the 25° microbridges is the occurrence of giant collective switching events over some volts, which can be explained by fast successive switching of adjacent Josephson junctions, due to interlayer coupling between these junctions, and a small spread of the critical currents. Our Tl-2212 microbridges can be modeled as a serial array of serial subarrays of intrinsic Josephson junctions. The length scale of the subarrays is in the range of some hundred nanometers. Since the parameter spread in such a subarray can be considered to be small, the measurement of such subarrays would be of great interest. The preparation of these subarrays is already planned. This will be achieved by narrowing the existing vicinal microbridges with the help of focused ion beam etching. Both the shunting effects of the 15° films as well as the collective switching behavior of the 25° films can be seen as a helpful condition for phase synchronization. However, the  $25^{\circ}$  films have more prospects for application (i.e., as local oscillators in the THz range) due to the smaller spread of critical currents, which is a good condition for phase synchronization as well as for the radiation of a strong collective signal. The next necessary step on this way is the preparation of single subarrays with a suitable technique, i.e., preparation with a focused ion beam.

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