NanoSQUIDs from YBa$_2$Cu$_3$O$_7$/SrTiO$_3$ superlattices with bicrystal grain boundary Josephson junctions

Jianxin Lin,$^1$ Julian Linek,$^1$ Reinhold Kleiner,$^1$ and Dieter Koelle$^1$

$^1$Physikalisches Institut, Center for Quantum Science (CQ) and LISA$^*$, University of Tübingen, Auf der Morgenstelle 14, 72076 Tübingen, Germany

(Dated: July 23, 2020)

We report on the fabrication and characterization of nanopatterned dc SQUIDs with grain boundary Josephson junctions based on heteroepitaxially grown YBa$_2$Cu$_3$O$_7$ (YBCO)/ SrTiO$_3$ (STO) superlattices on STO bicrystal substrates. Nanopatterning is performed by Ga focused-ion-beam milling. The electric transport properties and thermal white flux noise of superlattice nanoSQUIDs are comparable to single layer YBCO devices on STO bicrystals. However, we find that the superlattice nanoSQUIDs have more than an order of magnitude smaller low-frequency excess flux noise, with root-mean-square spectral density $S_{\phi}^{1/2} \sim 5 - 6 \mu_0 \Phi_0/\sqrt{Hz}$ at 1 Hz ($\Phi_0$ is the magnetic flux quantum). We attribute this improvement to an improved microstructure at the grain boundaries forming the Josephson junctions in our YBCO nanoSQUIDs.

PACS numbers: 85.25.CP, 85.25.Dq, 74.78.Na, 74.72.-h 74.25.F- 74.40.De

I. INTRODUCTION

Strongly miniaturized direct current (dc) superconducting quantum interference devices (SQUIDs) with dimensions in the submicrometer range (nanoSQUIDs) are promising devices for the sensitive detection and investigation of small spin systems, as they can be used for direct detection of the magnetization reversal of individual magnetic nanoparticles (MNP), nanotubes or nanowires$^{11,12}$ and for high-resolution scanning SQUID microscopy$^{11,12}$. The vast majority of nanoSQUIDs developed during the last decade are based on metallic superconductors, for which their operation temperature is limited by their transition temperature $T_c$ to below $\sim 10$ K. Furthermore, metallic low-$T_c$ superconductors typically have upper critical fields $B_{c2}$ below 1 T, which also limits the magnetic field range in which nanoSQUIDs based on them can be operated$^{12,13}$. As an alternative, the high-$T_c$ cuprate superconductor YBa$_2$Cu$_3$O$_7$ (YBCO), with $T_c \sim 92$ K and $B_{c2}$ well above 10 T offers the use of YBCO nanoSQUIDs for applications within a much wider range of temperature $T$ and magnetic field $B$, as compared to nanoSQUIDs based on conventional metallic superconductors.

The fabrication of Josephson junctions (JJ) and SQUIDs based on cuprate superconductors is demanding, in particular due to the very small superconducting coherence length on the nanometer scale and concomitant high sensitivity of those materials to defects on the atomic scale. YBCO micro- and nanoSQUIDs are typically based on single layers of epitaxially grown films, that are eventually covered with a metallic layer (e.g. Au) for resistive shunting and protection during nanopatterning$^{11}$. Josephson junctions in such single layer devices are based on constrictions (cJJs) with widths down to $\sim 50$ nm$^{22}$, that yield a root-mean-square (rms) spectral density of flux noise $S_{\phi}^{1/2}$ in the thermal white noise limit down to $< 450 \Phi_0/Hz^{1/2}$ at $T = 18$ K ($\Phi_0$ is the magnetic flux quantum$^{23}$). Another recently developed JJ type is based on creating Josephson barriers in YBCO thin films via focused ion beam (FIB) irradiation with a He ion beam$^{24}$. Sub-micron wide He-FIB JJ$^{25}$ and nanoSQUIDs$^{26}$ have been realized, and a rms flux noise in the thermal white noise limit down to $< 500 \Phi_0/Hz^{1/2}$ has been achieved at $T = 4.2$ K$^{27}$. Furthermore, grain boundary (GB) Josephson junctions (GBJJs), that are formed by epitaxial growth of YBCO on a bicrystal substrat$^{28}$ provide a well established approach for realizing YBCO nanoSQUIDs in single layer devices. The use of Ga FIB milling enables one to fabricate GBJJs down to 80 nm width$^{29}$, and operation of GBJJ based nanoSQUIDs up to $B = 3$ T$^{30}$ has been demonstrated. The optimization of the SQUID layout$^{31}$ led to the demonstration of ultralow rms flux noise in the thermal white noise limit down to $\sim 45 \Phi_0/Hz^{1/2}$ at $T = 4.2$ K$^{32}$. For a MNP placed in 10 nm-distance to a constriction in the SQUID loop, this corresponds to a spin sensitivity of $\sim 3 \mu_B/Hz^{1/2}$ ($\mu_B$ is the Bohr magneton). Such YBCO nanoSQUIDs with bicrystal GBJJs have been used for the investigation of the magnetization reversal of Fe and Co nanowires$^{27,33}$ and Co MNP$^{34}$ including e.g. the analysis of MNP switching fields over a wide $T$ range from 300 mK up to 80 K.

A significant drawback for high-$T_c$ cuprate SQUIDs in general is the very large amount of low-frequency excess noise, typically scaling with frequency $f$ as $S_{\phi} \propto 1/f$ (1/f noise)$^{35}$. The dominant source of 1/f noise comes from critical current $I_0$ fluctuations due to localized defects in the JJ barrier$^{36}$, which can be several orders of magnitude larger than for conventional Nb tunnel junctions$^{37,38}$. For YBCO nanoSQUIDs with ultralow thermal white noise, this excess noise can dominate the spectral density of flux noise up to high frequencies in the MHz range$^{39,40}$. An established procedure to suppress this 1/f noise contribution from $I_0$ fluctuations in dc SQUIDs is the application of bias reversal schemes.
(including proper modulation of flux bias), which suppresses 1/f noise below the applied bias reversal frequency $f_b$. However, applying such schemes to YBCO nanoSQUIDs is a particular challenge for the SQUID readout electronics and nanoSQUID design, because very large $f_b$ is required, and the required modulation of the flux bias points at $f_b$ is difficult to achieve due to the small SQUID inductance $L$ and concomitantly small mutual inductance $M$ between a nanoSQUID and the flux modulation line.

We further note that the flux noise spectrum of YBCO nanoSQUIDs often shows a superposition of a few Lorentzians rather than a pure 1/f spectrum, which can be explained by the relatively small number of dominating defects in JJs with small size. In addition we find excess 1/f noise in our GBJJ YBCO nanoSQUIDs, which has been attributed to the fluctuations of spins of unknown origin, most likely due to defects at the substrate/YBCO interface or at the edges of the nanopatterned structures. Another open issue is the unclear origin of strong 1/f noise in YBCO nanoSQUIDs based on cJJs. For those devices, this excess noise has been also attributed to $I_0$ fluctuations, although, there is no barrier in such nanowire.

Obviously, the strong low-frequency excess noise in YBCO nanoSQUIDs has to be related to the defect structure in the GB barrier or in the YBCO films forming cJJs, together with possible contributions from the YBCO/substrate interface or damaged edges. Apart from optimizing thin film growth by introducing suitable buffer or cap layers, a possible solution to this problem may be the growth of multilayers (superlattices), involving epitaxially grown interlayers between YBCO films to interrupt the growth of extended defects in YBCO. Such a multilayer or superlattice approach has been successfully used to significantly enhance the critical current density in YBCO films, in particular for thick films developed for high-current applications. Accordingly, the focus within this approach was on basic studies and on the improvement of pinning properties of YBCO films, often by deliberately introducing a high density of defects. Studies on 1/f noise in YBCO thin films and SQUIDs based on them, also indicated possible improvements via improved pinning properties.

However, so far, there is no information on the possible modification (improvement or deterioration) of 1/f noise from $I_0$ fluctuations in YBCO JJs and SQUIDs based on them, induced by the implementation of superlattice structures into the devices.

In this work, we report on the fabrication and properties of YBCO nanoSQUIDs that are based on a heteroepitaxial YBCO/STO superlattice, grown on a STO bicrystal substrate. The comparison with single layer YBCO devices of similar geometry shows, that the superlattice nanoSQUIDs yield comparable electric transport properties and comparable upper bounds to the flux noise in the thermal white noise limit. Regarding low-frequency excess noise, however, we find a strong reduction of the low-frequency noise in the superlattice nanoSQUIDs by more than one order of magnitude in rms flux noise at 1 Hz.

II. DEVICE FABRICATION AND LAYOUT

We first fabricated very thin c-axis oriented epitaxial single layer YBCO thin films by pulsed laser deposition (PLD) on (100)-oriented SrTiO$_3$ (STO) single crystal substrates. For films of thickness $d_f = 7, 17, 23$ and 31 nm, we find $T_c = 61, 81, 83$ and 84 K, respectively (via inductive $T_c$ measurements). Based on those results, we decided to fabricate multilayer (superlattice) devices with $d_f \approx 30$ nm per YBCO layer, to ensure a high enough $T_c$.

We used PLD to grow an epitaxial YBCO/STO superlattice on a STO bicrystal substrate with a symmetric (±12°) [001]-tilt grain boundary (GB), i.e., with a misorientation angle 2θ = 24° ($\theta = 12°$). The multilayer consists of a stack of four c-axis oriented YBCO layers with $d_f \approx 30$ nm thickness per layer, separated by three STO interlayers with 3 nm thickness per layer. The choice of the total YBCO thickness $d = 4d_f \approx 120$ nm is based on an optimization study, that revealed optimum spin sensitivity for this choice of $d$.

For all layers, we used the same deposition parameters (substrate temperature $T_s = 800°C$ and oxygen partial pressure $P_O$, 0.2 mbar). For details of the growth process of our YBCO thin films on STO and of their structural and electric transport properties see e.g. Refs. [30, 48, and 49]. An in-situ evaporated 65-nm-thick Au layer on top of the YBCO/STO superlattice serves as a resistive shunt to provide nonhysteretic current-voltage characteristics (IVCs) of the grain boundary junctions formed in YBCO and as a protection layer during Ga FIB nanopatterning.

For the characterization of the crystalline quality of the unpatterned YBCO/STO multilayer, we performed x-ray diffraction (XRD) using a 4-circle PhilipsX’Pert diffractometer. Fig. 1(a) shows a $\theta$–2$\theta$ scan, indicating single phase c-axis oriented YBCO films. The $\omega$ scan (rocking curve), shown in the inset of Fig. 1(a), yields good alignment of the c-axis perpendicular to the substrate plane, with full-width-half-maximum (FWHM) $\sim 0.16^\circ$. From the position of the YBCO (005) peak we extract a c-axis lattice constant of 11.69 A, which is close to the value of fully oxygenated unstrained YBCO [24]. Fig. 1(b) shows a YBCO (103) $\varphi$-scan with diffraction peaks at $0^\circ \pm (\theta)$, $90^\circ \pm (\theta)$, $180^\circ \pm (\theta)$ and $360^\circ \pm (\theta)$, reflecting the fourfold symmetry of the crystal lattice and rotation of the crystallographic axes by $\pm \theta$ across the GB. No other peaks can be detected, which confirms in-plane epitaxial growth just according to the orientation of the STO bicrystal without misaligned grains.

After characterization of the unpatterned YBCO/STO multilayer, we used photolithography and Ar ion milling to prepattern 16 microbridges straddling the grain boundary to create 8 µm wide
FIG. 1. XRD data of the unpatterned YBCO/STO superlattice on a STO bicrystal substrate. (a) $\Theta - 2\Theta$ scan, showing YBCO (00ℓ) Bragg peaks. Inset shows rocking curve of the (005) peak. (b) YBCO (103) plane XRD $\varphi$-scan. Black and red indicate diffraction peaks from the two grains of the bicrystal.

GBJJs; those will be referred to as JJ-1 to JJ-16. Those bridges are connected to several-mm long and few-100$\mu$m wide contact pads onto which Al wires are bonded as voltage and current leads for 4-point measurements. Subsequently, YBCO nanoSQUIDs with similar sizes are nanopatterned into some of the 8$\mu$m-wide GBJJs by FIB milling with 30-keV Ga ions in a dual-beam FIB system.

Fig. 2 shows scanning electron microscopy (SEM) images of two YBCO/STO multilayer nanoSQUIDs SQ-14 and SQ-15, nanopatterned into JJ-14 and JJ-15, respectively. At the position where the GB crosses the microbridge, the Ga FIB is used to define two sub-$\mu$m-wide GBJJs with width $w_{JJ}$ ($i = 1, 2$) and to mill the SQUID hole with width $\sim \ell_J \times \ell_c$. In addition, we use the Ga FIB to cut a slit perpendicular to the GB (from the right in Fig. 2) towards the GB, to produce a constriction with width $w_c$ in the SQUID loop. By applying a modulation current $I_{\text{mod}}$ through the constriction (indicated as arrows in Fig. 2(a)), the magnetic flux $\Phi$ in the SQUID can be modulated, to ensure SQUID operation at optimum flux bias and to perform SQUID readout in a flux-locked loop (FLL). For measurements of magnetization reversal of individual MNPs, placing the MNP on top of the constriction also provides optimum coupling of signal to the nanoSQUID.

On JJ-15 we determined from resistance vs $T$ measurement $T_c = 87.2$ K, with a transition width $\Delta T_c \approx 0.8$ K. For SQ-15 (nanopatterned into JJ-15), we determined essentially unchanged $T_c$ and $\Delta T_c$ after FIB patterning.

III. TRANSPORT AND NOISE PROPERTIES OF SUPERLATTICE JJS AND NANOSQUIDS

In this section, we present and discuss electric transport and noise properties measured in liquid Helium at $T = 4.2$ K in an electrically and magnetically shielded environment. We focus on two fabricated YBCO/STO superlattice nanoSQUIDs, SQ-14 and SQ-15, which were FIB-patterned from JJ-14 and JJ-15, respectively. For comparison we also show data for a single layer (SL) 120 nm-thick YBCO nanoSQUID (SQ-SL), also with a 65 nm-thick Au layer on top, with comparable lateral geometry. This SL nanoSQUID was FIB-patterned from the 8$\mu$m-wide JJ-SL, also on a STO bicrystal substrate with misorientation angle $2\theta = 24^\circ$.

The IVCs of JJ-14 and JJ-15, measured before FIB patterning, are shown in Fig 3. Those were recorded in zero applied magnetic field ($H = 0$). The IVCs have
FIG. 3. IVCs (at $T = 4.2$ K and $H = 0$) of 8-µm-wide GB-JJs in a YBCO/STO superlattice (JJ-14 and JJ-15) and in a single layer YBCO film (JJ-SL), all with Au on top.

the typical shape described by the resistively and capacitively shunted junction (RCSJ) model without hysteresis. We note that the 8-µm-wide JJs are in the long junction limit, as also revealed by $I_c(H)$ measurements (not shown). The $I_c(H)$ curves show a slight asymmetry, which means that the maximum critical current $I_{c,\text{max}}$ is not reached exactly at $H = 0$; this is the reason for the slight asymmetries in the IVCs (different critical current for opposite polarity). Hence, the $I_{c,\text{max}}$ values quoted in Table I have been obtained from the maxima of the $I_c(H)$ patterns. Values of $I_{c,\text{max}}$ for both JJs are around 3 mA. Converting this into a critical current density $j_c$ by dividing by the JJ width (8 µm) and total YBCO film thickness (120 nm) yields $j_c \approx 3 \times 10^5$ A/cm$^2$ (see also Table I). This is a typical value which we obtain for YBCO GBJJs of similar size, made from single layer films and hence indicates, that the current flowing through the JJs is distributed across all four YBCO layers in the superlattice. This observation is also consistent with microstructural studies on YBCO/STO superlattices with 3 nm-thick STO layers.

With a measured resistance $R_n \sim 0.1$ Ω (which is dominated by the shunting Au layer), for the two JJs, we obtain a characteristic voltage $V_c = I_{c,\text{max}}R_n \sim 0.3$ mV (see also Table I); this is at the lower end of the range of values of $V_c$ that we observe for single layer YBCO nanoSQUIDs based on GBJJs on STO bicrystal substrates. For comparison, the IVC of JJ-SL is also shown in Fig. 3; this device indeed has similar values for $I_c$, $R_n$ and $V_c$; c.f. Table I. We note that values of $I_{c,\text{max}}$ and $R_n$ for the other 8-µm-wide JJs on the same chip with the YBCO/STO superlattice are very similar to JJ-14 and JJ-15. 

Fig. 4 shows IVCs of superlattice nanoSQUIDs SQ-14 and SQ-15 and, for comparison, the IVC of the single layer nanoSQUID SQ-SL, both for bias current polarities.

FIG. 4. IVCs of superlattice SQUIDs (SQ-14 and SQ-15) and single layer SQUID (SQ-SL). Solid (dashed) lines are recorded with $I_{\text{mod}}$ adjusted to obtain maximum (minimum) critical current $I_{c,\text{max}}$ for $I > 0$. 

FIG. 5. Critical current $I_c$ vs. $I_{\text{mod}}$ of multilayer SQUIDs (SQ-14 and SQ-15) and single layer SQUID (SQ-SL) for both bias current polarities.

FIG. 5. Critical current $I_c$ vs. $I_{\text{mod}}$ of multilayer SQUIDs (SQ-14 and SQ-15) and single layer SQUID (SQ-SL) for both bias current polarities.

With a measured resistance $R_n \sim 0.1$ Ω (which is dominated by the shunting Au layer), for the two JJs, we obtain a characteristic voltage $V_c = I_{c,\text{max}}R_n \sim 0.3$ mV (see also Table I); this is at the lower end of the range of values of $V_c$ that we observe for single layer YBCO nanoSQUIDs based on GBJJs on STO bicrystal substrates. For comparison, the IVC of JJ-SL is also shown in Fig. 3; this device indeed has similar values for $I_c$, $R_n$ and $V_c$; c.f. Table I. We note that values of $I_{c,\text{max}}$ and $R_n$ for the other 8-µm-wide JJs on the same chip with the YBCO/STO superlattice are very similar to JJ-14 and JJ-15. 

With a measured resistance $R_n \sim 0.1$ Ω (which is dominated by the shunting Au layer), for the two JJs, we obtain a characteristic voltage $V_c = I_{c,\text{max}}R_n \sim 0.3$ mV (see also Table I); this is at the lower end of the range of values of $V_c$ that we observe for single layer YBCO nanoSQUIDs based on GBJJs on STO bicrystal substrates. For comparison, the IVC of JJ-SL is also shown in Fig. 3; this device indeed has similar values for $I_c$, $R_n$ and $V_c$; c.f. Table I. We note that values of $I_{c,\text{max}}$ and $R_n$ for the other 8-µm-wide JJs on the same chip with the YBCO/STO superlattice are very similar to JJ-14 and JJ-15. 

With a measured resistance $R_n \sim 0.1$ Ω (which is dominated by the shunting Au layer), for the two JJs, we obtain a characteristic voltage $V_c = I_{c,\text{max}}R_n \sim 0.3$ mV (see also Table I); this is at the lower end of the range of values of $V_c$ that we observe for single layer YBCO nanoSQUIDs based on GBJJs on STO bicrystal substrates. For comparison, the IVC of JJ-SL is also shown in Fig. 3; this device indeed has similar values for $I_c$, $R_n$ and $V_c$; c.f. Table I. We note that values of $I_{c,\text{max}}$ and $R_n$ for the other 8-µm-wide JJs on the same chip with the YBCO/STO superlattice are very similar to JJ-14 and JJ-15. 

With a measured resistance $R_n \sim 0.1$ Ω (which is dominated by the shunting Au layer), for the two JJs, we obtain a characteristic voltage $V_c = I_{c,\text{max}}R_n \sim 0.3$ mV (see also Table I); this is at the lower end of the range of values of $V_c$ that we observe for single layer YBCO nanoSQUIDs based on GBJJs on STO bicrystal substrates. For comparison, the IVC of JJ-SL is also shown in Fig. 3; this device indeed has similar values for $I_c$, $R_n$ and $V_c$; c.f. Table I. We note that values of $I_{c,\text{max}}$ and $R_n$ for the other 8-µm-wide JJs on the same chip with the YBCO/STO superlattice are very similar to JJ-14 and JJ-15.
TABLE I. Summary of geometric and electric parameters of 8 μm-wide JJs and nanoSQUIDs as defined in the text. JJ-14 and JJ-15 are based on YBCO/STO superlattices, from which SQ-14 and SQ-15 was nanopatterned, respectively. JJ-SL is based on a single layer of YBCO, from which SQ-SL was nanopatterned. For all devices the total YBCO thickness is 120 nm and the Au layer on top is 65 nm thick.

| Device | \( w_{c1} \) (nm) | \( w_{c2} \) (nm) | \( t_c \) (nm) | \( \ell_c \) (nm) | \( I_{c,\text{max}} \) (mA) | \( R_n \) (Ω) | \( V_c \) (mV) | \( j_c \) (10^5 A/cm²) | \( I_{\text{mod,0}} \) (mA) | \( M \) (pH) | \( L \) (pH) | \( \beta_L \) |
|--------|----------------|----------------|-------------|----------------|----------------|-------|--------|----------------|----------------|-------|-------|-------|
| JJ-14  | 3.2            | 0.091          | 0.29        | 3.3            | 3.3            |       | 3.3    |               |                |       |       |       |
| JJ-15  | 3.3            | 0.098          | 0.32        | 3.4            | 3.4            |       | 3.4    |               |                |       |       |       |
| JJ-SL  | 3.4            | 0.07           | 0.23        | 3.5            | 3.5            |       | 3.5    |               |                |       |       |       |
| SQ-14  | 280            | 320            | 180         | 250            | 500            | 0.42  | 0.82   | 0.34          | 5.9            | 0.26  | 0.90  | 2.3   | 7.1   | 1.5   |
| SQ-15  | 250            | 280            | 280         | 200            | 400            | 0.29  | 1.28   | 0.37          | 4.5            | 0.12  | 2.19  | 1.0   | 4.9   | 0.7   |
| SQ-SL  | 280            | 290            | 300         | 350            | 400            | 0.20  | 1.68   | 0.34          | 3.0            | 0.12  | 1.24  | 1.7   | 12    | 1.2   |

a value of \( M \) inbetween. This observation is consistent with the different values for the constriction width \( w_c \) (cf. Table I and Fig. 2). We find that a narrower constriction yields a larger \( M \). This is also supported by inductance calculations based on simulations of the supercurrent density distribution in our nanoSQUIDs via the software 3D-MLSI, which solves the London equation in 2-dimensional current sheet [44][52]. A larger \( M \) is beneficial, as it relaxes requirements (maximum feedback and modulation currents) on the SQUID readout electronics for FLL operation and as it also improves the coupling between a MNP and a nanoSQUID, resulting in improved spin sensitivity.

To obtain a rough estimate of the inductance \( L \) of the nanoSQUIDs, we determine the screening parameter \( \beta_L \equiv 2LI_0/\Phi_0 \) from the modulation depth \( \Delta I_c \equiv I_{c,\text{max}} - I_{c,\text{min}} \) of the \( I_c(I_{\text{mod}}) \) oscillations. Into the definition of \( \beta_L \) enters the noise free critical current \( I_0 \) of the JJs. In the case of negligible noise rounding, as applicable to our devices at 4.2 K (cf. Fig. 2), we can replace \( 2I_0 \) by \( I_{c,\text{max}} \). Then, by using the dependence \( \Delta I_c/I_{c,\text{max}}(\beta_L) \) derived from numerical simulations for symmetric dc SQUIDs in the noise-free case, we determine \( \beta_L \) values as listed in Table I and which are close to the value \( \beta_L \approx 1 \) for optimum flux noise [57][58]. From the estimated values for \( \beta_L \) and the measured values for \( I_{c,\text{max}} \) we then obtain the values for \( L \) as listed in Table I. For the superlattice nanoSQUIDs we obtain values of \( L \) below 10 pH. Achieving such a small inductance is important for obtaining very low values of flux noise for the nanoSQUIDs in the thermal white noise limit well below \( S_{\Phi}^{1/2} = 1 \mu \Phi_0/Hz^{1/2} \).

We note that all \( I_c(I_{\text{mod}}) \) curves in Fig. 5 show a clear asymmetry. This is visible as a shift of the patterns along the \( I_{\text{mod}} \) axis in opposite direction for opposite current polarities and a skewness of the \( I_c(I_{\text{mod}}) \) curves, which can arise from asymmetries in the critical currents of the two JJs and from inductance asymmetry [57][58]. These observations are consistent with the slight asymmetry in the widths of the JJs, inducing a critical current asymmetry, and with the fact that the constrictions in the SQUIDs induce an inductance asymmetry.

Fig. 6 shows \( V(I_{\text{mod}}) \) oscillations of SQ-15, measured at different bias currents. The shift of those curves along the \( I_{\text{mod}} \) axis for opposite bias current polarity is consistent with the concomitant shift in the \( I_c(I_{\text{mod}}) \) pattern for SQ-15 shown in Fig. 5. Moreover, the maxima in \( V(I_{\text{mod}}) \) are slightly shifting along the \( I_{\text{mod}} \) axis with increasing bias current, which is consistent with a small inductance asymmetry, as discussed above. We only show here \( V(I_{\text{mod}}) \) oscillations for SQ-15; however, the same features are also present for SQ-14 and SQ-SL.

Finally, we discuss the flux noise of the superlattice YBCO nanoSQUIDs, which has been measured in FLL mode. Fig. 7(a) shows the rms spectral density of flux noise \( S^{1/2}(f) \) of SQ14 and SQ-15 measured with dc bias and with bias reversal at frequency \( f_{\text{br}} = 20 \) kHz. In the bias reversal mode, the contribution of critical current fluctuations to low-frequency excess noise is removed below \( f_{\text{br}} \). From the estimated values for \( \beta_L \) and the measured values for \( I_{c,\text{max}} \) we then obtain the values for \( L \) as listed in Table I. For the superlattice nanoSQUIDs we obtain values of \( L \) below 10 pH. Achieving such a small inductance is important for obtaining very low values of flux noise for the nanoSQUIDs in the thermal white noise limit well below \( S_{\Phi}^{1/2} = 1 \mu \Phi_0/Hz^{1/2} \).

We note that all \( I_c(I_{\text{mod}}) \) curves in Fig. 5 show a clear asymmetry. This is visible as a shift of the patterns along the \( I_{\text{mod}} \) axis in opposite direction for opposite current polarities and a skewness of the \( I_c(I_{\text{mod}}) \) curves, which can arise from asymmetries in the critical currents of the two JJs and from inductance asymmetry [57][58]. These observations are consistent with the slight asymmetry in the widths of the JJs, inducing a critical current asymmetry, and with the fact that the constrictions in the SQUIDs induce an inductance asymmetry.

Fig. 6 shows \( V(I_{\text{mod}}) \) oscillations of SQ-15, measured at different bias currents. The shift of those curves along the \( I_{\text{mod}} \) axis for opposite bias current polarity is consistent with the concomitant shift in the \( I_c(I_{\text{mod}}) \) pattern for SQ-15 shown in Fig. 5. Moreover, the maxima in \( V(I_{\text{mod}}) \) are slightly shifting along the \( I_{\text{mod}} \) axis with increasing bias current, which is consistent with a small inductance asymmetry, as discussed above. We only show here \( V(I_{\text{mod}}) \) oscillations for SQ-15; however, the same features are also present for SQ-14 and SQ-SL.

Finally, we discuss the flux noise of the superlattice YBCO nanoSQUIDs, which has been measured in FLL mode. Fig. 7(a) shows the rms spectral density of flux noise \( S^{1/2}(f) \) of SQ14 and SQ-15 measured with dc bias and with bias reversal at frequency \( f_{\text{br}} = 20 \) kHz. In the bias reversal mode, the contribution of critical current fluctuations to low-frequency excess noise is removed below \( f_{\text{br}} \). We note that we do not reach the thermal white noise regime even at the highest frequency of 100 kHz up to which we performed measurements. Hence the rms flux noise values \( S^{1/2}(100 \) kHz) = 244 n\( \Phi_0/Hz^{1/2} \) for
substrates, as reported very recently.

for single layer YBCO nanoSQUIDs on MgO bicrystal
tal substrates

so far for single layer YBCO nanoSQUIDs on STO bicrys-
than an order of magnitude lower than what we obtained

This observation shows, that the use of YBCO/STO su-
perlattices instead of YBCO single layer films can signifi-
cantly reduce 1/f noise in YBCO nanoSQUIDs based on

GbJJs.

The noise spectra for SQ-14 and SQ-15 in bias revers-
mode yield a significant improvement of $S_\phi^{1/2}(1\text{ Hz})$
over dc bias readout, for SQ-15 even below $1\mu\Phi_0/\text{Hz}^{1/2}$
[cf. Fig. 7(a)]. This shows, that $I_0$ fluctuators in the
GBJJ barriers are the major source of low-frequency
excess noise in our devices, stemming from defects in
the barriers. Hence, we conclude that the significantly
reduced low-frequency excess noise in the YBCO/STO
superlattice nanoSQUIDs is most likely due to an
improved quality of the grain boundary, as compared to
single layer YBCO nanoSQUIDs. Finally, we note that
we still see low-frequency excess noise even with bias re-
versal readout. This issue has been addressed already in
Ref. [32] where we attributed this to possible contribu-
tions from fluctuating spins in the substrate close to the
STO/YBCO interface.

IV. CONCLUSIONS

We have fabricated YBCO dc nanoSQUIDs from a
YBCO/STO superlattice, consisting of four 30 nm-thick
individual YBCO layers, separated by 3 nm-thick STO
layers. The superlattice is grown heteroepitaxially on a
STO bicrystal substrate with 24° misorientation angle,
and covered with 65 nm-thick Au on top, as a resistive
shunt and for protection during Ga FIB milling.
The characterization of crystalline film quality and measure-
ment of the electric transport properties before and after
Ga FIB nanopatterning shows that the superlattice de-
vice have comparable quality as for single layer devices
with the same total YBCO film thickness. Also the mea-
sured noise properties in the thermal white noise limit are
similar for single layer and superlattice devices. This is
in strong constrast to the observed low-frequency excess
noise: superlattice devices yield more than one order of
magnitude lower noise at 1 Hz as compared to single layer
devices.

Because the low-frequency excess noise is dominated
by fluctuations of the critical current $I_0$ in the grain
boundary Josephson junctions, we attribute the im-
proved low-frequency noise performance of the superlat-
ce nanoSQUIDs to an improved microstructure of the
grain boundaries forming the Josephson junctions. How-
ever, so far we have no direct information on the mi-
crostructure at the grain boundaries in our superlattices.
One possible reason for the improvement might be due to
reduced facetting of the grain boundaries. Certainly, a
clarification of this issue, or of other possible modifica-
tions in the defect structure of the grain boundaries, that
are induced by inserting STO interlayers in the YBCO
films, is required in future studies.

In any case, the achieved improvement in low-
frequency excess noise in YBCO nanoSQUIDs provides
the opportunity to realize ultrasensitive devices for scan-
ning SQUID microscopy and for the investigation of

FIG. 7. Rms spectral density of flux noise $S_\phi^{1/2}(f)$ measured
in FLL mode: (a) data for SQ-14 and SQ-15, measured with
dc bias and bias reversal ($f_{dc} = 20\text{ kHz}$). (b) dc bias readout
data of SQ-14, SQ-15, together with SQ-SL and SQ-SL2 on
an expanded scale for $S_\phi^{1/2}$ and $f$. 

SQ-14 and 104 $\mu\Phi_0/\text{Hz}^{1/2}$ for SQ-15 (with dc bias read-
out) are upper bounds for the thermal white noise limit.
Those are comparable to the best values for the flux noise
at high frequencies obtained for YBCO nanoSQUIDs
based on GBJJs in single layer devices [32,50].

Most importantly, with dc bias readout we obtain at
$f = 1\text{ Hz}$ values $S_\phi^{1/2}(1\text{ Hz}) = 6.5\mu\Phi_0/\text{Hz}^{1/2}$
for SQ-14 and 4.9 $\mu\Phi_0/\text{Hz}^{1/2}$ for SQ-15. Those values are more
than an order of magnitude lower than what we obtained
so far for single layer YBCO nanoSQUIDs on STO bicry-
tal substrates [32,50] and they are comparable to the values
for single layer YBCO nanoSQUIDs on MgO bicrystal
substrates, as reported very recently [30]. To illustrate this
observation, we show in [2, b] again the rms flux noise
measured in dc bias mode for SQ-14 and SQ-15, now to-
gether with noise data for the single layer devices SQ-SL
and SQ-SL2. The latter has also been fabricated on an
STO bicrystal substrate with an Au layer on top. SQ-SL2 has
been reported earlier to show the so far lowest flux
noise of $\sim 45\mu\Phi_0/\text{Hz}^{1/2}$ in the thermal white noise limit
(at very high frequency $>7\text{ MHz}$) for any of our YBCO
nanoSQUIDs [32]. The flux noise at $f = 1\text{ Hz}$ for both sin-
gle layer devices shown in Fig. 7(b) is $\sim 80\mu\Phi_0/\text{Hz}^{1/2}$.
This observation shows, that the use of YBCO/STO su-
perlattices instead of YBCO single layer films can signifi-
cantly reduce 1/f noise in YBCO nanoSQUIDs based on
GbJJs.
magnetization reversal processes in individual magnetic nanosystems, by utilizing already achievable ultralow levels of white noise and expand those on the frequency scale down to well below the MHz range. Moreover, the superlattice approach may also be helpful to improve the low-frequency noise performance of other devices, e.g. sensitive SQUID magnetometers, that are based on grain boundaries. It remains to be shown, whether this superlattice approach could also improve the low-frequency excess noise in YBCO nanoSQUIDs based on nanowires (constriction junctions).

CONFLICTS OF INTEREST

The authors declare no conflicts of interest

ACKNOWLEDGMENTS

J. Lin acknowledges funding by the China Scholarship Council (CSC). We gratefully acknowledge fruitful discussions and technical support by M. Turad and R. Löffler (LISA+) and by C. Back. This work was supported by the COST action NANOCOHYBRI (CA16218).

1 W. Wernsdorfer, “Classical and quantum magnetization reversal studied in nanometerized particles and clusters,” Adv. Chem. Phys. 118, 99–190 (2001)
2 M. Jamet, W. Wernsdorfer, C. Thirion, D. Mailly, V. Dupuis, P. Méline, and A. Pérez, “Magnetic anisotropy of a single cobalt nanocluster,” Phys. Rev. Lett. 86, 4676–4679 (2001)
3 W. Wernsdorfer, “From micro- to nano-SQUIDs: applications to nanomagnetism,” Supercond. Sci. Technol. 22, 064013 (2009)
4 L. Hao, C. Altmann, J. C. Gallop, D. Cox, F. Ruede, O. Kazakova, P. Josephs-Franks, D. Drung, and Th. Schurig, “Detection of single magnetic nanobead with a nano-superconducting quantum interference device,” Appl. Phys. Lett. 98, 092504 (2011)
5 A. Buchter, J. Nagel, D. Rüffer, F. Xue and D. P. Weber, O. F. Kieler, T. Weismann, J. Kohlmann, A. B. Zorin, E. Russo-Averchi, R. Huber, P. Berberich, A. Fontcuberta i Morral, M. Kemmler, R. Kleiner, D. Koelle, D. Grundler, and M. Poggio, “Reversal mechanism of an individual Ni nanotube simultaneously studied by torque and SQUID magnetometry,” Phys. Rev. Lett. 111, 067202 (2013)
6 C. Granata and A. Vettoliere, “Nano superconducting quantum interference device: A powerful tool for nanoscale investigations,” Phys. Rep. 614, 1–69 (2016)
7 M. J. Martínez-Pérez and D. Koelle, “NanoSQUIDs: Basics & recent advances,” Phys. Sci. Rev. 2, 20175001 (2017)
8 M. J. Martínez-Pérez, B. Müller, D. Schweins, D. Korinski, R. Kleiner, J. Sesé, and D. Koelle, “NanoSQUID magnetometry of individual cobalt nanoparticles grown by focused electron beam induced deposition,” Supercond. Sci. Technol. – focus issue on NanoSQUIDs and their applications 30, 024003 (2017)
9 M. J. Martínez-Pérez, J. Pablo-Navarro, B. Müller, J. Stankievicz, R. Kleiner, C. Magén, D. Koelle, J. M. de Teresa, and J. Sesé, “NanoSQUID magnetometry on individual as-grown and annealed Co nanowires at variable temperature,” Nano Lett. 18, 7674–7682 (2018)
10 M. J. Martínez-Pérez, B. Müller, J. Lin, L. A. Rodriguez, E. Snoeck, R. Kleiner, J. Sesé, and D. Koelle, “Magnetic vortex nucleation and annihilation in bi-stable ultra-small ferromagnetic particles,” Nanoscale 12, 2587–2595 (2020)
11 J. R. Kirtley, M. B. Ketchen, K. G. Stavrias, J. Z. Sun, W. J. Gallagher, S. H. Blanton, and S. J. Wind, “High-resolution scanning SQUID microscope,” Appl. Phys. Lett. 66, 1138–1140 (1995)
12 J. R. Kirtley, “Prospects for imaging magnetic nanoparticles using a scanning SQUID microscope,” Supercond. Sci. Technol. 22, 064008 (2009)
13 D. Vasyukov, Y. Anahory, L. Embon, D. Halbertal, J. Cuppens, L. Ne’eman, A. Finkler, Y. Segev, Y. Myasoedov, M. L. Rappaport, M. E. Huber, and E. Zeldov, “A scanning superconducting quantum interference device with single electron spin sensitivity,” Nature Nanotechnol. 8, 639–644 (2013)
14 D. Koelle, “Magnetic sensors: A tip for better sensing,” Nature Nanotechnology 8, 617–618 (2013)
15 D. Hazra, J. R. Kirtley, and K. Hasselbach, “Nano-superconducting quantum interference devices with continuous read out at millikelvin temperatures,” Appl. Phys. Lett. 103, 093109 (2013)
16 J. R. Kirtley, L. Paulius, A. J. Rosenberg, J. C. Palmstrom, C. M. Holland, E. M. Spanton, D. Schiessl, C. L. Jermain, J. Gibbons, Y.-K.-K. Fung, M. E. Huber, D. C. Ralph, M. B. Ketchen, G. W. Gibson Jr., and K. A. Moler, “Scanning SQUID susceptometers with sub-micron spatial resolution,” Rev. Sci. Instr. 87, 093702 (2016)
17 P. Reith, X. R. Wang, and H. Hilgenkamp, “Analysing magnetism using scanning SQUID microscopy,” Rev. Sci. Instr. 88, 123706 (2017)
18 M. Wyss, S. Gliga, D. Vasyukov, L. Ceccarelli, G. Romagnoli, J. Cui, A. Kleibert, R. L. Stamps, and M. Poggio, “Stray-field imaging of a chiral artificial spin ice during magnetization reversal,” ACS Nano 13, 13910–13916 (2019)
19 Y. Anahory, H. R. Naren, E. O. Lachman, S. Buibut Sinai, A. Uri, L. Embon, E. Yaakobi, Y. Myasoedov, M. E. Huber, R. Klajn, and E. Zeldov, “SQUID-on-tip with single-electron spin sensitivity for high-field and ultra-low temperature nanomagnetic imaging,” Nanoscale 12, 3174–3182 (2020)
20 A. Uri, Y. Kim, K. Bagani, C. K. Lewandowski, S. Grover, N. Auerbach, E. O. Lachman, Y. Myasoedov, T. Taniguchi,
ramp-zigzag Josephson junctions," [Phys. Rev. B 86, 144531 (2012)].

50 J. Lin, B. Müller, J. Linek, M. Karrer, M. Wenzel, M. J. Martínez-Pérez, R. Kleiner, and D. Koelle, “YBa$_2$Cu$_3$O$_7$ nano superconducting quantum interference devices on MgO bicrystal substrates,” [Nanoscale 12, 5658–5668 (2020)].

51 J. D. Jorgensen, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus, and W. K. Kwok, “Structural properties of oxygen-deficient YBa$_2$Cu$_3$O$_{7-\delta}$,” [Phys. Rev. B 41, 1863–1877 (1990)].

52 W. C. Stewart, “Current-voltage characteristics of Josephson junctions,” [Appl. Phys. Lett. 12, 277–280 (1968)].

53 D. E. McCumber, “Effect of ac impedance of dc voltage-current characteristics of Josephson junctions,” [J. Appl. Phys. 39, 3113–3118 (1968)].

54 L. Ryen, E. Olsson, C. N. L. Edvardsson, and U. Helmersson, “Dislocations, strain, and defects in heteroepitaxial YBa$_2$Cu$_3$O$_{7-\delta}$/SrTiO$_3$ multilayers,” [Physica C 304, 307–313 (1998)].

55 M. M. Khapaev, M. Y. Kupriyanov, E. Goldobin, and M. Siegel, “Current distribution simulation for superconducting multi-layered structures,” [Supercond. Sci. Technol. 16, 24–27 (2003)].

56 J. Nagel, O. F. Kieler, T. Weimann, R. Wölbinger, J. Kohlmann, A. B. Zorin, R. Kleiner, D. Koelle, and M. Kemmler, “Superconducting quantum interference devices with submicron Nb/HfTi/Nb junctions for investigation of small magnetic particles,” [Appl. Phys. Lett. 99, 032506 (2011)].

57 C. D. Tesche and J. Clarke, “DC SQUID: Noise and optimization,” [J. Low Temp. Phys. 29, 301–331 (1977)].

58 B. Chesca, R. Kleiner, and D. Koelle, “SQUID Theory,” in The SQUID Handbook, Vol. I: Fundamentals and Technology of SQUIDs and SQUID systems, edited by John Clarke and Alex I. Braginski (Wiley-VCH, Weinheim, 2004) Chap. 2, pp. 29–92.