Epitaxial thick film high-$T_c$ SQUIDs

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Abstract. Low-noise operation of superconducting quantum interference devices (SQUIDs) in magnetic fields requires high critical current and strong pinning of vortices in the superconducting electrodes and in the flux transformer. Crack-free epitaxial high-$T_c$ dc-SQUID structures with a total thickness $\sim 5 \mu m$ and a surface roughness determined by 30 nm high growth spirals were prepared with YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) films on MgO substrates buffered by a SrTiO$_3$/BaZrO$_3$-bilayer. HRTEM demonstrated a high quality epitaxial growth of the films. The YBCO films and SQUID structures deposited on the buffered MgO substrates had a superconducting transition temperature $T_c$ exceeding 91 K and critical current densities $J_c > 3 \text{ MA/cm}^2$ at 77 K up to a thickness $\sim 5 \mu m$. The application of thicker superconducting and insulator films helped us to increase the critical current and dynamic range of the multilayer high-$T_c$ flux transformer and improve the insulation between the superconducting layers. An optimization of SQUID inductance allowed us to fabricate 8 mm SQUID magnetometers with SQUID voltage swings of $\sim 60 \mu V$ and a field resolution of $\sim 30 \text{ fT/}\sqrt{\text{Hz}}$ at 77 K.

1. Introduction
Magnetometers based on superconducting quantum interference devices (SQUIDs) are the most sensitive sensors for measurements of magnetic field components (see, e.g., [1, 2]). An increase of the operation temperature up to 77 K and the possibility of performing measurements without magnetic shielding can significantly reduce costs and broaden the field of applications. Present day flip-chip high-$T_c$ dc-SQUID magnetometers have demonstrated a field resolution better than $4 \text{ fT/}\sqrt{\text{Hz}}$ at 77 K in magnetic shielding [3, 4]. In the case of unshielded operation, the SQUIDs are exposed to the Earth’s magnetic field, where the detected field component can change during SQUID movements from $-50 \mu T$ to $+50 \mu T$, and line-frequency electromagnetic fields. The latter vary from $\sim 10 \text{ nT}$ up to several $\mu T$ in peak-to-peak swing under laboratory conditions at different locations in the building. In combination with the few $\text{fT/}\sqrt{\text{Hz}}$ field resolution of the SQUIDs such conditions impose strong requirements on the dynamic range and the slew rate of the sensitive high-$T_c$ dc-SQUID systems. In addition to a perforated or slotted layout of the SQUID washer [5, 6], the pinning of Abrikosov vortices in the superconducting films of the SQUID body should be improved to reduce low-frequency noise of the SQUIDs in magnetic fields. The SQUID voltage swing should be also enhanced for a larger slew rate and frequency bandwidth of the measurement system. In the present paper we describe high-$T_c$ flip-chip dc-SQUID magnetometers optimized for the sensitive measurements without magnetic shielding in the Earth’s magnetic field.
2. Experimental

BaZrO$_3$ (BZO), SrTiO$_3$ (STO), YBa$_2$Cu$_3$O$_{7-x}$ (YBCO), and PrBa$_2$Cu$_3$O$_{7-x}$ (PBCO) films were deposited on symmetric 20$^\circ$ or 24$^\circ$ bicrystal or single crystal MgO substrates [7] by the high-oxygen-pressure dc-sputtering technique [8]. The STO/BZO-bilayer was used as an epitaxial buffer on the MgO substrates. The thermal expansion coefficients of MgO and YBCO are very similar and this has allowed us to produce up to $\sim$5 µm thick crack-free high-quality YBCO films and YBCO-PBCO structures. The deposition rate of the YBCO and PBCO films was $\sim$50 nm/hour while the deposition rate of the non-conducting materials was $\sim$10 nm/hour. To reduce mechanical strain in the structures and for a better oxygenation of the thick YBCO films the samples were slowly heated (0.1 deg/min) up to 700 $^\circ$C in a flow of pure (99.999%) molecular oxygen followed by 0.1 deg/min cooling to room temperature.

The thickness of the films was measured by a surface profiler Dektak IIA. The surface morphology of the films and structures was verified by optical and atomic force microscopy (AFM). The quality of the crystal structure of the films was investigated by high resolution transmission electron microscopy (HRTEM) with a spatial resolution better than 0.1 nm. The critical current density $J_c$ of the YBCO films was estimated by the non-destructive permanent magnet method [9] and for YBCO films with a thickness < 300 nm it was checked by 4-point measurements of the IV characteristics of patterned bridges with 1 µV criteria. The PBCO films were used in ramp-type junctions and for the insulation layer in crossovers of the multilayer flux transformers [10]. Patterning of the structures was performed by ion beam etching with the exception of the non-aqueous Br-ethanol etching of the first two layers of the ramp junctions and the multilayer flux transformers. AZ5214 and AZMIR701 photoresists were used for structuring with ion beam etching while Br-ethanol etching was performed with a PMMA mask [10].

![Figure 1. Optical image of the surface of the 3 µm thick YBCO film with transition temperature $T_c = 93$ K.](image1)

![Figure 2. HRTEM image of the YBCO/STO interface obtained in the [110] direction.](image2)
BZO/MgO and STO/BZO interfaces in the buffer layers of our structures were recently published [11]. There it was shown that the BZO buffer layer at the BZO/MgO interface provided the epitaxial growth of the top layer films with perovskite structures (STO, YBCO, and PBCO) on the rock-salt structure of MgO, while the matching of the lattice constants appeared by relaxation of the atomic structure at the STO/BZO interface. As a demonstration of the perfect epitaxial growth of YBCO films on the STO buffer layer we present here an HRTEM image of the interface between STO and YBCO films. It was determined (see Fig. 2) that the interface atomic layer has a Ba$_{1-x}$Sr$_x$O composition so it belongs to both STO and YBCO layers.

The high and constant critical current density throughout the thickness of the thick YBCO films is a real challenge and important for all applications of high-T$_c$ superconductors. The addition of non-superconducting nanoprecipitates of BZO or Y$_2$BaCuO$_5$ was suggested and successfully applied to increase the $J_c$ of YBCO films [12, 13]. On the other hand, it is already well known [8] that the YBCO films deposited by high oxygen pressure sputtering technique naturally contain lattice-coherent non-superconducting Y$_2$O$_3$ nanoparticles of optimal size and separation [4, 13]. The Y$_2$O$_3$ nanoparticles are nearly spherical with a diameter of $\sim$20 nm and are homogeneously distributed with a separation of $\sim$30 nm [4]. Similar to the findings of Kim et al. [13], a strong 3D pinning of the Abrikosov vortices is expected in the bulk of the thick films with a uniform local $J_c$ $\sim$3.5 MA/cm$^2$. Fortunately, we are able to produce much thicker YBCO films without cracks or thickness degradation of the current-carrying cross section in the films, which is demonstrated, for example, by the good surface morphology of the thick films (see Fig. 1).

The standard procedure for measuring the critical current density of the films by structuring narrow bridges is not convenient for thick films. The required currents are in the ampere range and can heat the sample at the contacts for the current inputs. A more practical and fast estimation of the critical current density can be done by a non-destructive permanent magnet method suggested in [9]. We measured the dependence of the repulsive force “F” between a 6 mm diameter permanent SmCo$_5$ magnet and an YBCO film of thickness “d”. The magnet brought to a distance of $\sim$1 mm from the YBCO film, while the latter was immersed in liquid nitrogen at 77 K. A linear dependence F(d) was observed up to a film thickness of $\sim$5 µm (see Fig.3).

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**Figure 3.** Dependence of the maximum repulsive force “F” between the 6 mm SmCo$_5$ magnet and YBCO films vs. the film thickness “d”.

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The linear dependence $F(d)$ corresponds to a thickness-independent $J_c \sim 3.4 \text{ MA/cm}^2$ according to the empirical relationship $[9] J_c \sim 0.17 \left(\frac{F}{d}\right) \text{[MA/cm}^2\text{]},$ where $F$ is the maximum repulsive force measured in gram weight and $d$ is the film thickness in micrometers. For a small film thickness $d \leq 300 \text{ nm}$ this $J_c$ evaluation was additionally calibrated with a conventional direct 4-point measurement of the critical current.

The currents induced in the thick YBCO films were able to shield the $\sim 1 \text{ kG}$ strong magnetic field of the SmCo$_5$ magnet. At 77 K 1 cm wide strips of a 4.5 µm thick film have an estimated total critical current of $\sim 1.5 \text{ kA},$ which exceeds the critical current of the present day 2$^{nd}$ generation high-T$_c$ superconducting tapes of similar width about 15-fold (see, e.g., [12, 14] and references therein).

High-quality thick and flat YBCO films are an important prerequisite for the preparation of present dc SQUID sensors. We used the thick ($> 1 \mu$m) YBCO films for the preparation SQUIDs with ramp-type or bicrystal Josephson junctions and multilayer flux transformers. In the case of ramp-type Josephson junctions, the thick top electrode shields the junction area thus improving the stability of operation in magnetic fields. A photograph and noise spectra of a 3 µm thick dc SQUID with ramp-type Josephson junctions are shown in Fig. 4 and Fig. 5, respectively. It was observed that an increase of the low-frequency noise appeared first at magnetic fields above 300 µT while in the case of the SQUIDs with standard 100 nm thick electrodes this already takes place at a magnetic field of $\sim 100 \mu$T [5].

![Figure 4](image1.png)  
**Figure 4.** Photograph of the first 3 µm thick SQUID with the ramp-type Josephson junctions.

![Figure 5](image2.png)  
**Figure 5.** Noise spectra in the magnetic fields for the 3 µm thick SQUID with the ramp-type Josephson junctions.

Bicrystal grain boundary (GB) Josephson junctions with the required critical current have a fixed cross-section determined by the misorientation angle, but the wide electrodes of the junctions can be made much thicker (see Fig. 7). This should reduce the demagnetizing coefficient of the wider parts of the SQUID body and provide a better pinning of the Abrikosov vortices to reduce the low-frequency noise of the SQUIDs in the magnetic field.

![Figure 7](image3.png)  
**Figure 7.** Schematic representation of the grain boundary (GB) Josephson junction with the variable thickness YBCO thick film electrodes.
We integrated such “variable thickness” GB junctions in the flip-chip dc-SQUID magnetometers and gradiometers with multilayer flux transformers similar to those described in [15]. The typical thickness of the YBCO film of the SQUID washer was \( \sim 1.5 \mu m \). The total thickness of the multilayer flux transformers was \( \sim 3 \mu m \). The critical current of the flux transformer \( I_c \) \( \sim 0.3 A \) at 77 K was limited by the cross-section area of the lines in the input coil of \( \sim 10 \mu m^2 \). In the case of 8 mm flux transformers for the HTM-8-type flip-chip magnetometers [15], this current provided a maximal dynamic range of about \( \pm 190 \mu T \) while in the case of 16 mm flux transformers of the HTM-16-type magnetometers the maximal dynamic range was about \( \pm 100 \mu T \). These values are sufficient to follow changes of the magnetic field during movement of the sensors in the Earth’s magnetic field. The thick SQUID washer has withstood the changes of the magnetic field amplified by the flux transform.

For the HTM-8-type magnetometers described in [15] an estimated SQUID inductance \( L_s \sim 80 \text{ pH} \), typical SQUID voltage swing \( V_{pp} \sim 30 \mu V \), a spectral density of the flux noise \( \Phi \) \( \Phi / \sqrt{Hz} \), and a field sensitivity \( B_N \sim 15 \text{ fT/Hz} \) at 77 K were achieved. We have observed that a reduction of the SQUID inductance down to \( L_s \sim 45 \text{ pH} \) for the new HTM-8x-type magnetometers increased the voltage swing up to \( V_{pp} \sim 60 \mu V \), reduced flux noise to \( \Phi / \sqrt{Hz} = 9 \mu \Phi / \sqrt{Hz} \) but increased the field noise up to \( B_N \sim 30 \text{ fT/Hz} \) at 77 K. The estimated SQUID inductance of the HTM-8x-type magnetometers was \( L_s \sim 50 \text{ pH} \), consisting of \( \sim 40 \text{ pH} \) of geometrical inductance and \( \sim 10 \text{ pH} \) kinetic inductance [16]. The kinetic inductance was reduced down to \( \sim 3 \text{ pH} \) by the increased thickness of the YBCO film in the SQUID washer. This was observed by an increase of the voltage swing for the magnetometers with similar layouts but with different thickness of the YBCO film in the dc-SQUID washer. The energy sensitivity of the HTM-8x sensors is \( \varepsilon = S_{\Phi} / 2L_s \sim 3 \times 10^{-30} \text{ J/Hz} \) at 77 K. The dc-SQUID sensors were operated in an unshielded environment without suppression of either the bias current or the SQUID voltage swing. The white noise of the SQUID sensors in unshielded environment was not changed either.

3. Discussion

The technology of epitaxial thick film high-T_c SQUID magnetometers developed here is the main advance described in the present paper. A better reproducibility of the flip-chip sensors and their low-noise operation in magnetic fields was achieved. An additional optimization of the SQUID inductance has resulted in doubling of the SQUID voltage swing with a simultaneous reduction of the flux noise. The frequency bandwidth and slew rate are important for the operation of unshielded SQUID systems. Both of them are inversely proportional to the system flux noise \( S_{\Phi} \) (see, e.g., [17]) which, in turn, is proportional to the square of SQUID inductance [18]:

\[
S_{\Phi} = S_T \left( \frac{\partial V}{\partial \Phi} \right)^2 \approx \left( 4k_B T R_D^2 / R_D \right) \left[ 1 + \frac{1}{2} \left( \frac{I_0}{I} \right)^2 \right] \left( R_n / L_s \right)^2 \propto \frac{L_s^2}{R_n} \tag{1}
\]

A slew rate of \( \sim 5 \text{ MfT/s} \) was demonstrated for the HTM-8 magnetometers with direct readout SQUID electronics [19]. For the new HTM-8x-type magnetometers with the SQUID inductance \( L_s \sim 50 \text{ pH} \) the expected slew rate is \( \sim 10 \text{ MfT/s} \), which is a typical slew rate of the systems with the low-T_c SQUID magnetometers that have a similar voltage swing [20].

The field resolution of the inductively coupled magnetometers should improve with reduction of \( L_s \) as long as there is no significant degradation of the coupling coefficient “k” [18]:

\[
B_N = \frac{L_p + L_i}{k A \sqrt{L_i L_s}} S_{\Phi}^{1/2} \propto \frac{1}{k} \sqrt{L_s} \propto \frac{L_s}{R_n} \tag{2}
\]
where for the HTM-8x-type flip-chip high-Tc dc-SQUID magnetometers the inductance of the pick-up loop \( L_p \sim 40 \text{ nH} \) is equal to the inductance of the input coil \( L_i \) and the area of the pick-up loop \( A \sim 64 \text{ mm}^2 \). Equations 1 and 2 can explain qualitatively our experimental observations if we take into account that the coupling coefficient “k” decreases at smaller \( L_S \).

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