Long-time stable high-temperature superconducting DC-SQUID gradiometers with silicon dioxide passivation for measurements with superconducting flux transformers

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Abstract
In applications for high-$T_c$ superconducting DC-SQUIDs such as biomagnetism, nondestructive evaluation and the relaxation of magnetic nanoparticles, it is important to maintain reliable sensor performance over an extended time period. We have designed and produced DC-SQUID gradiometers based on YBa2Cu3O7−x (YBCO) thin films which are inductively coupled to a flux transformer to achieve a higher sensitivity. The gradiometers are protected against ambient atmosphere and humidity by SiO2 and amorphous YBCO layers.

The noise properties of the sensor in flip-chip configuration, especially in unshielded environments, are shown. We present a comparison of Tl2Ba2CaCu2O8+x (TBCCO) thin films on buffered sapphire or LaAlO3 substrates for the flux transformer in shielded and unshielded environments. We reach a low white field gradient noise of 72 fT cm$^{-1}$ Hz$^{-1}$ with the TBCCO on LaAlO3 flux transformer. The electric properties of the gradiometers (critical current $I_C$, normal state resistance $R_N$ and the transfer function $V_\Phi$) were measured over a period of one year and do not show significant signs of degradation.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
Long-term sensor stability is a requirement for the application of high-temperature superconducting DC-SQUID gradiometers. Changes in the electrical characteristics of sensors lead to decreased performance and a higher cost of the final system. The most sensitive areas of the gradiometer are the Josephson junctions, which we created using single-layer bicrystal technology. An overview of the properties of grain boundary Josephson junctions is given in [1]. A possible cause for degradation of superconducting thin films is the diffusion of oxygen in the YBCO layer. In monocrystalline films the oxygen transport occurs along the a–b plane of the crystals [2]. At growth defects or grain boundaries a significant oxygen diffusion appears along the c-axis of the epitaxially grown film [3, 4]. Therefore a passivation at the sides and on top of the superconducting thin film is necessary to reduce diffusion. We developed a process to compensate for height differences during
fabrication and protected our sensors using different materials to achieve stability over a period of more than one year.

2. Fabrication

The sensors described here are galvanically coupled DC-SQUID gradiometers (for layout see figure 1). The YBCO thin films, having a thickness of 150 nm, are grown along the c-axis on SrTiO3 bicrystal substrates with pulsed laser deposition [5]. The thin films are patterned with Ar-ion beam etching and planarized in situ via the deposition of 150 nm amorphous YBCO with RF-sputtering. For repeated measurements with different combinations of readout gradiometers and flux transformers the sensors need to be protected against mechanical damage and require high-quality bond contacts. The direct contact between the readout gradiometer and the flux transformer in the flip–chip package can lead to damages in the thin films which influence the electrical properties of the system. A protective layer is also necessary to reduce the diffusion of oxygen and to avoid contact between the superconducting thin film and environmental humidity. In our current set-up we use SiO2 for this layer on the top of the YBCO film. Further details regarding the thin film system are given in [6].

We have developed a new technique to prepare the bonding pads of the sample. The aim was to improve the adhesion of the bond wire and to lower the contact resistance. Therefore, prior to the YBCO deposition, we grew a 50 nm thin gold layer on the substrate using a lift-off technique. During heating of the substrate to the deposition temperature of 750 °C the gold layer clusters into small islands with a diameter of 1–2 μm (see figure 2). Between these islands the YBCO growth is well c-oriented with a rocking-curve width of less than 0.3° and a critical temperature higher than 86 K. The contact resistance of an average bonding pad was lowered from 160 mΩ (without gold clusters) to <4 mΩ (with gold clusters).

The SQUID consists of two Josephson junctions with a width of 3 μm and a galvanic coupling to the antenna structures. The pick-up loops of the antenna structure are used in a gradiometric configuration. An external magnetic field gradient leads to a difference in magnetic flux in the antenna structures. This difference is galvanically coupled into the SQUID with the current ΔI (see figure 3) and an inductance of the incoupling line of the SQUID loop $L_M$. The length of the SQUID loop is either $l = 50 \mu m$ or in a second layout $l = 70 \mu m$ (see figure 3), the width of the incoupling lines is 5 μm, and the inner hole area is 4 μm wide. In former investigations we have shown that, for a film thickness of about 150 nm, the inductance $L_M$ of a DC-SQUID with a width of the incoupling lines of 5 μm and an inner area of 3 μm can be estimated by a factor of 1.05 pH μm$^{-1}$ times the loop length. The decrease of the width of incoupling lines to 4 μm and the increase of the width of the inner area to 5 μm increase this factor to a value of about 1.8 pH μm$^{-1}$ [7]. With these specifications we estimate the coupling inductance of the DC-SQUIDs in our gradiometers to be in the range of 50 pH, in the ideal case, to 90 pH for a loop length of 50 μm. The exact value depends on the accuracy of the photolithography and patterning process. To determine the coupling inductance we usually measure the flux modulation of the DC-SQUID with a current directly coupled into the SQUID loop [8]. This precise method is only possible after disconnecting the antenna structures so that these measurements can only be made after long-term experiments.
Figure 4. Flip–chip configuration with readout gradiometer on the left side and flux transformer on the right side. The difference in screening currents $\Delta I_f$ is magnetically coupled into the readout gradiometer.

To optimize the coupling between the antenna of the readout gradiometer and the antenna of the flux transformer we used different widths $W_{ant}$ for the antenna of the readout gradiometer (200, 400 and 600 $\mu$m; see figure 1) [9].

The flux transformer was prepared on a separate substrate (see figure 4). We used two different substrate materials and TBCCO as the superconducting thin-film material. The comparison between YBCO and TBCCO on different substrate materials for the flux transformer has shown that TBCCO leads to the best sensor performance, i.e., the lowest white noise level, due to a better flux pinning in an unshielded environment [10]. The size of the complete flux transformer is $8 \times 40$ mm$^2$ and the baselength $b_f$ is 19 mm. After patterning by wet etching, a 300 nm layer of SiO$_2$ is deposited on the flux transformer as protection against chemical and mechanical influences. The two pick-up loops of the flux transformer are connected in parallel. The current caused by a difference in magnetic flux is inductively coupled to the readout gradiometer with an S-shaped superconducting line, which is optimized to the dimensions of the readout gradiometer.

3. Electrical characteristics

We focused our measurements on the electrical characteristics at liquid nitrogen temperatures. The sensors were evaluated over a period of one year in a magnetically shielded environment (two layers of $\mu$-metal) to reduce the influence of external disturbances. During this period each sensor was cooled and then heated to room temperature ten times to simulate environmental stress during the normal sensor lifetime. Between the measurements the gradiometers and flux transformers were stored at room temperature with a humidity of 40–45%.

To measure their $I–V$ characteristics, the DC-SQUID gradiometers were biased with different currents and the resulting voltage signal was amplified and recorded. With this information we calculated the critical current $I_C$, normal state resistance $R_N$, and the $I_C R_N$ product (see figure 5). The $I_C R_N$ product allows one to compare different types of Josephson junctions independent of the junction geometry. This parameter should change significantly if degradation between the measurements occurs. To evaluate the response to external magnetic fields we calculated the transfer function of the sensors over time (see figure 6). At a fixed bias current the maximum voltage modulation $V_{pp}$ is measured as a function of the current in a small copper coil, $I_M$. Assuming a sinusoidal $V(I_M)$ characteristic the transfer function $V_\phi$ can be calculated as $V_\phi = \pi V_{pp}$.

4. Experimental results

An important improvement to realize sensors with a long lifetime was the usage of SiO$_2$ passivation layers and amorphous YBCO films. As a first step we examined whether there is a negative influence of the passivation on the superconducting properties of the sensors. With all tested sensors the SiO$_2$ thin film did not change the electrical characteristics, and the dielectric influence is negligible compared to the influence of the substrate material (SrTiO$_3$).

The planarization of the structures with amorphous YBCO deposited by hollow cathode sputtering does not lead to a large change of the superconducting properties of bicrystal Josephson junctions. In some cases we observed a slight increase of the critical current, compared to bicrystal junctions.
Long-time stable HTSC DC-SQUID gradiometers with flip–chip flux transformers

Figure 7. Field gradient resolution of the readout gradiometer without and with the flux transformer.

without planarization. This can be caused by oxygen diffusion between the planarization layer consisting of YBCO$_{7-x}$ with $x \approx 0$ and the crystalline thin film with $0.1 > x > 0$. An increase of the critical current is primarily observed for bicrystal angles of $30^\circ$ and above.

During the processing of the gradiometer, especially during the dry-etching and photolithography steps, a temperature increase of the thin film up to $100^\circ$C accelerates the diffusion of oxygen into the area of the Josephson junctions instead of out of this area as observed without amorphous YBCO planarization thin films. To lower the rate of diffusion out of the antenna areas, the substrate is cooled by liquid nitrogen during the dry-etching procedure [11, 12].

The comparison among gradiometers (see figure 5) shows a large spread of the $I_C R_N$ product due to different current densities of the grain boundary junctions used for these experiments. All of these junctions show the typical scaling behaviour for grain boundary junctions [10]. Compared to previous experiments by other groups, only the $I_C R_N$ value of 30 $\mu$V for the $30^\circ$ grain boundary junction (curve F) is relatively low [13]. The experimental investigations of the long-term stability shows no relevant changes or signs of degradation in most cases (see figure 5). Only small changes of the $I_C R_N$ product were measured. In all of these cases the reason for this behaviour was a decrease of the respective critical current. A decreasing $I_C$ does not automatically lead to a decrease in the transfer function, as ageing processes can change $R_N$ at the same time.

The $24^\circ$ and $30^\circ$ grain boundary junctions used in our experiments exhibit relatively high critical current densities. In combination with a width of the SQUID structures in the range of 3 $\mu$m the critical currents can be as high as 100 $\mu$A. With high critical currents the inductance parameter $\beta_L$ can exceed the optimum value ($\leq 1$), which leads to a decreasing transfer function [8, 14]. Higher $\beta_L$ values reduce the voltage modulation due to a screening of the flux which is coupled into the SQUID. Therefore a slight decrease of the critical current results in an improvement of the transfer function (sample B and E). Such a decrease can be caused by diffusion during the processing of the superconducting thin film or by slower diffusion during normal operation of the sensor.

In the case of increasing critical current (sample G) we see a reduction of the transfer function $V_d$ in figure 6. In our experiments the transfer function increased significantly for only one sample with a $30^\circ$ grain boundary (sample F); the reason for this is topic of future investigations.

The fabricated gradiometers display a large spread in the transfer function with a minimum of 10 $\mu$V. However, all sensors were able to operate in an unshielded environment with commercially available SQUID electronics (Magnicon, Philips).

In the next step we have measured various combinations of readout gradiometers and flux transformers in shielded as well as unshielded environments [10]. The field gradient resolution $\sqrt{S_G}$ is proportional to the flux noise $\sqrt{S_{\Phi_1}}$ and depends on the effective area of the configuration $A_{\text{eff}}$ and the base length $b$:

$$\sqrt{S_G} = \frac{\sqrt{S_{\Phi_1}}}{b A_{\text{eff}}}$$

(1)

The responsitivity of a gradiometer is defined as the product of effective area and base length. Figure 7 shows as an example the increase of the field gradient resolution in an unshielded environment for the readout gradiometer (sample G) in comparison to the flip–chip configuration. This increase is caused by the large flux transformer in the flip–chip configuration with a higher base length ($b_f \approx 1.9$ cm) and an increased effective area ($A_{\text{eff}} \approx 0.22$ mm$^2$).

The field gradient resolution as a function of the responsitivity for different readout gradiometers at 1 Hz and in the white noise region is illustrated in figure 8. By changing the width of the readout gradiometer antenna $w_{\text{ant}}$ the

Figure 8. Field gradient resolution over responsitivity in a magnetically unshielded environment with readout gradiometer on the left-hand side and flip–chip configuration on the right-hand side.
responsivity can be adjusted, but the field gradient resolution is mostly unchanged. The reason for this behaviour is the higher flux noise in an unshielded environment for larger antenna widths. In particular, the largest in-coupling inductance $L_{in}$ (loop length $l = 70 \mu m$) together with the largest antenna width results in the largest responsivity ($>0.06 \text{ cm mm}^2$, $\phi$), but leads to a relatively low field gradient resolution in an unshielded environment, caused by the large flux noise of $200 \mu \Phi_0 \text{ Hz}^{-1/2}$ in comparison to the lower value of about $50 \mu \Phi_0 \text{ Hz}^{-1/2}$ for the other gradiometer layouts.

This means that, for the development of sensors in the flip–chip configuration, it can be helpful to optimize the readout gradiometers in the direction of low flux noise in an unshielded environment, not in the direction of higher responsivity. As shown in figure 8 we realized the best value in the field gradient resolution with the readout gradiometer with the lowest level in the flux noise ($<50 \mu \Phi_0 \text{ Hz}^{-1/2}$, $l = 50 \mu m$, $200 \mu m$ line width, $\phi$) in an unshielded environment not with the sensor with the higher responsivity ($0.04 \text{ cm mm}^2$). If the readout gradiometer alone has a large responsivity ($A_{eff}$) the additional flux transformer only improves the field gradient resolution by a factor of 2.

With a similar flux noise, it should be possible to reduce the field gradient resolution significantly due to the higher responsivity of the combination of readout gradiometer and flux transformer. We are able to achieve a reduction by a factor of 5, corresponding to a field gradient resolution of $880 \text{ fT cm}^{-1}, \text{ Hz}^{-1/2}$ at frequencies around 1 Hz and a reduction by a factor of 20 ($72 \text{ fT cm}^{-1}, \text{ Hz}^{-1/2}$) at higher frequencies, showing the effectiveness of this approach.

5. Summary

The developed and fabricated sensors were successfully protected by a combination of an amorphous YBCO and SiO$_2$ layers against oxygen diffusion. After a period of one year no significant signs of degradation were detected.

With TBCCO flux transformers on LaAlO$_3$ substrates stacked with readout gradiometers, we achieve a field gradient noise of $72 \text{ fT cm}^{-1}, \text{ Hz}^{-1/2}$ for the white noise level and $880 \text{ fT cm}^{-1}, \text{ Hz}^{-1/2}$ at 1 Hz in unshielded environments. We are continuing to investigate the stability over longer time periods and include the noise investigations in shielded as well as unshielded environments.

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