Development of high sensitive magnetic contaminant detection system using an HTS-rf-SQUID covered with HTS thin films

Y Hatsukade, R Kurosawa, Y Uchida, and S Tanaka
Department of Environmental & Life Sciences, Toyohashi University of Technology, 1-1 Hibarigaoka, Tenpaku-cho, Toyohashi, Aichi 441-8580, Japan
E-mail: hatukade@ens.tut.ac.jp

Abstract. For high sensitive detection of magnetic contaminant in electrode of lithium-ion battery, high-temperature superconductor (HTS) radio-frequency (rf) superconducting quantum interference devices (SQUIDs) based on a bicrystal SrTiO$_3$ (STO) substrate was designed, and fabricated employing YBa$_2$Cu$_3$O$_{7-x}$ thin films of about 200 nm in thickness. To improve characteristics such as effective area and 1/f noise profile of the SQUID, HTS thin films on normal STO substrates were overlapped on a wide superconducting weak link and/or a slit of the SQUID in flip-chip configuration. The noise profiles of the SQUID covered with the films on the respective positions were well improved compared to that of the bare SQUID. A magnetic contaminant detection system was developed employing the HTS-rf-SQUID covered with the films on both the positions. Using this system, a tungsten ball of 30 μm in diameter was successfully detected with a signal to noise ratio of about 14.

1. Introduction
Detection of ultra-small magnetic contaminants is a crucial issue for manufactures producing commercial products such as electrodes of lithium ion batteries. Although the industry is requiring a technology to identify the fine magnetic particles of less than 50 μm in diameter in the products, X-ray imaging and eddy current testing, which are commonly employed as the detection method, are unable to detect such small objects when the products are moving in the production lines. Therefore, a practical and high sensitive detection system for fine magnetic contaminants is demanded.

In this study, we designed and fabricated an YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) high temperature superconductor (HTS) radio frequency (rf) superconducting quantum interference device (SQUID) with a rectangular hole based on a bicrystal SrTiO$_3$ (STO) substrate as a magnetic sensor [1, 2]. The HTS-rf-SQUID based on the single-layer YBCO thin-film technology and the bicrystal Josephson junction was employed to take advantage of the high magnetic sensitivity, high time resolution, the simple fabrication process, and easy-handling of the SQUID operated in liquid nitrogen. The characteristics of the HTS-rf-SQUIDs such as effective area and 1/f noise profile were improved by covering the SQUID partially with HTS thin films in a flip-chip configuration. We developed a magnetic contaminant detection system utilizing the HTS-rf-SQUID covered with the HTS films and the characteristics and the performance of the system were investigated.

2. Design, fabrication and evaluation of HTS-rf-SQUID
The pattern of the single-layer HTS rf SQUIDs based on the bicrystal STO substrate used in this study is shown in Figure 1. Concerning the HTS rf SQUID with an inductance $L$, resistance $R$, and critical
current $I_c$, when it is operated with SQUID electronics with a tank circuit in the so-called adiabatic regime, where the resonance frequency $\omega_{rf}$ is much smaller than both the junction characteristic frequency $\omega_c = 2\pi R I_c / \Phi_0$ and the cut-off frequency of the SQUID ring $\omega_{cut} = 2\pi R I / L$, and in the hysteretic mode, where the hysteretic parameter $\beta_{rf} = 2\pi L I / \Phi_0 > 1$ and the noise parameter $\Gamma = 2\pi k_B T(I, \Phi_0) \ll 1$, the following relation holds between the SQUID inductance $L$ and the flux-to-voltage transfer function $dV/d\Phi$ with an appropriate quality factor $Q$ of the tank circuit [3]:

$$
\frac{dV}{d\Phi} \approx \frac{\omega_{rf} L_r}{M} = \frac{\omega_{rf}}{k} \sqrt{\frac{L_r}{L}}
$$

where $M$ and $k$ are the mutual inductance and the coupling coefficient between $L$ and $L_r$ of the tank circuit. In order to achieve a lower flux noise level, $dV/d\Phi$ must be larger. $dV/d\Phi$ is inversely proportional to the root of $L$, while $L$ is determined by the hole shape in the case of a washer SQUID ring [4]. In this study, we employed the rectangular shape of 800 x 50 $\mu$m² for the hole, since the SQUID inductance with a rectangular hole is smaller than that with a square hole with the same area. The outer dimension of the SQUID was 9 x 9 mm².

We fabricated the HTS-rf-SQUIDs based on the design shown in Figure 1. YBCO thin film with a thickness of about 200 nm was deposited on a bicrystal STO substrate with a mis-orientation angle of 30°. By conventional photolithography and ion-etching technologies, the SQUID with the bicrystal Josephson junctions of 4 $\mu$m in width was patterned. The SQUID had superconducting weak link with width of about 4 mm on the bicrystal grain boundary (GB) due to the SQUID pattern.

The fabricated HTS-rf-SQUID was operated with a SQUID electronics developed by Juelicher SQUID GmbH [5]. The SQUID was inductively coupled with a normal-conducting antenna from the electronics using the bicrystal STO substrates as the substrate resonator, which forms the tank circuit with the antenna [6]. The characteristics of the SQUID were measured in a magnetically shielded room (MSR) with a shielding factor of about 60 dB while cooling the SQUID in liquid nitrogen. The resonance frequencies $\omega_{rf}$ was about 680 MHz. The magnetic field noise profile of the SQUID is shown in Figure 2. In the measurements, the data was measured three times, and then averaged. The white flux and field noise levels $S_{\phi}^{1/2}$ and $S_{B}^{1/2}$ at 1 kHz were about 52 $\mu\Phi_0$/Hz$^{1/2}$ and 120 fT/Hz$^{1/2}$, respectively. The effective area $A_{eff}$ of the SQUID was about 0.81. We note that the white field noise level was sufficiently low although the 1/f noise arose at about a few hundreds Hz and was relatively high compared to those of HTS-rf-SQUIDs with step-edge Josephson junctions [6, 7].
3. HTS-rf-SQUID with HTS thin films in flip-chip configuration

In the SQUID pattern shown in Figure 1, the bicrystal GB runs across the washer SQUID ring to form the wide superconducting weak link, where penetration and movement of the flux vortices can easily occur [8]. For the application of the HTS-rf-SQUID to the magnetic contaminant detection, where the weak magnetic signal in the low frequency range less than 100 Hz must be measured, the reduction of the 1/f noise is strongly desirable. In order to prevent flux vortices from penetrating and moving in the wide weak link in the SQUID pattern, we fabricated HTS thin films to cover the weak link in a flip-chip configuration. YBCO thin film with the same thickness as the SQUID was deposited on a normal STO substrate without GB, and then rectangular pieces with an area of 4.1 x 2 mm$^2$ were cut out by a diamond saw. The cut YBCO films were positioned on the SQUID so that one of them covered the wide weak link while the other covered the slit of the SQUID. Both films were set carefully so as not to cover the SQUID hole. The positions of the films on the SQUID are indicated in the inset of Figure 3 with the dashed lines. The former film was expected to serve as a superconducting shield with its Meissner effect to avoid the penetration and motion of flux vortices in the weak link, while the latter was expected to serve as a superconducting shield to couple external magnetic flux to the SQUID hole, which would pass through the slit without the film, to increase the effective area. For adhesion between the films and the SQUID, we applied Apiezon grease between them, and pressed both the substrates so that the distance between the SQUID and the films became as small as possible. Polymer threads were used to reinforce the connection between the SQUID and the films tightly. We measured the characteristics such as the effective area $A_{\text{eff}}$ and the field noise $S_B^{1/2}$ of the following four cases: the bare HTS-rf-SQUID, the SQUID with the HTS film on the GB, the SQUID with the HTS film on the slit, and the SQUID with both films on the respective positions. The measurements were carried out in the same MSR while cooling the SQUID with or without the HTS films in liquid nitrogen. The resonance frequency of the SQUID with the films slightly changed from the original value of 680 MHz of the bare SQUID. All the measurement data were obtained three times with different HTS films, and then averaged.

The field noise profiles of the SQUID with the HTS films on the GB and/or the slit are shown in Figure 3. The noise profile of the bare SQUID mentioned above is shown by the dotted line for comparison. The characteristics of all the SQUIDs with and without the films are summarized in Table I. The SQUID with the film on the GB shows superior noise characteristics to the others, especially in the low frequency range of less than 10 Hz. On the other hand, the noises from 10 to 100 Hz of the SQUIDs with the film on the slit and the films on both positions were better than that with only the film on the GB. This must be due to the fact that the effective areas $A_{\text{eff}}$ of the formers were larger than that of the latter. The coverage of the slit contributed to increase the area by about 13% as expected. In addition, it is noteworthy that the 1/f noise was also reduced compared to the bare SQUID. It is inferred that the density of the external flux must be higher around the edges on the slit due to focusing effect of a HTS thin film. The decrease in the 1/f noise should be because the coverage of the film on the slit contributed to the reduction of the flux trapping on the edges. We also note that the coverage of the GB also contributed to increase the effective area moderately. As shown in ref. 9, the increase in the number of flux trapped in a HTS film indicates the increase in non-superconducting area of the film. Therefore, we think that the possible removal of flux trapping in the GB by the coverage with the HTS film may serve to recover the actual effective area of the SQUID. Not as expected, the 1/f noise in the low frequency range of less than 10 Hz in the case of the SQUID with the two films was highest among the SQUIDs with the films, although the noise profile is better than that of the bare SQUID. In this case, the SQUID hole was left uncovered by the films. Therefore, the large 1/f noise with the coverage of both the GB and the slit may be due to the flux trapping at the edges of the SQUID hole, where the external flux density must be higher.
4. Magnetic contaminant system using HTS-rf-SQUID covered with HTS thin films

We developed a magnetic contaminant detection system with the HTS-rf-SQUID covered with the HTS films on both the GB and the slit utilizing the SQUID microscopic technique. A block diagram of the system is depicted in Figure 4 (a). The system consists of the covered HTS-rf-SQUID in the microscopic cryostat, SQUID electronics, electromagnetic and magnetic shielded cases, permanent magnet, belt conveyers, filters and PC. In this system, an object under test including a fine magnetic contaminant is set on the belt conveyer, and then magnetized in the vertical direction by the permanent magnet of 0.25 T. After the magnetization, the object is conveyed under the SQUID, which was installed in the cryostat and is cooled at about 77 K conductively, at a velocity of 100 mm/s. The remnant magnetic signal from the magnetized contaminant is measured by the SQUID, and the SQUID output voltage is recorded through a high-pass filter with a cut-off frequency of 0.5 Hz and a low-pass filter with a cut-off frequency of 20 Hz in the PC. The schematic figure of the SQUID mounted in the cryostat is illustrated in Figure 4 (b). The SQUID with the films was fixed with silver past on the sapphire rod, which was connected to a copper tank filled with liquid nitrogen. In order to reduce the liftoff distance, the normal STO substrates with original thickness of 0.5 mm were manually filed such that the thickness became to be about 0.25 mm. A sapphire window with thickness of 0.5 mm separates the SQUID and the room temperature. The minimum liftoff distance between the SQUID to an object is estimated to be about 1 mm. Figure 5 shows the noise profile of the covered SQUID installed in the system. For comparison, the noise of the bare SQUID without the films in the

Table 1. Characteristics of HTS-rf-SQUID with and without HTS films on GB and slit at 77 K.

| SQUID condition      | \( A_{\text{eff}} \) [mm\(^2\)] | \( S_{B}^{1/2} \) [fT/Hz\(^{1/2}\)] @ 1 Hz | \( S_{B}^{1/2} \) [fT/Hz\(^{1/2}\)] @ 10 Hz | \( S_{B}^{1/2} \) [fT/Hz\(^{1/2}\)] @ 100 Hz |
|----------------------|-------------------------------|------------------------------------------|------------------------------------------|------------------------------------------|
| Bare SQUID           | 0.81                          | 1930                                     | 440                                      | 240                                      |
| Film on GB           | 0.83                          | 440                                      | 190                                      | 170                                      |
| Film on slit         | 0.92                          | 900                                      | 210                                      | 140                                      |
| Films on GB & slit   | 0.93                          | 2100                                     | 225                                      | 135                                      |
system is shown together. The noise level of the covered SQUID between 6 Hz to 100 Hz was lower than that of the bare SQUID.

By this system, detection of fine tungsten balls with diameters of less than 100 μm was examined. Figure 6 (a) shows the experimental results of a tungsten ball of 100 μm in diameter measured with the bare SQUID and the covered SQUID with the liftoff distances of about 1.5 and 1.75 mm, respectively. Due to the larger effective area, the signal measured with the covered SQUID was about 10 % larger than that measured with the bare SQUID, even though the liftoff distance of the former was a bit longer than that of the latter. The smallest tungsten ball of 30 μm in diameter was successfully detected with a signal to noise ratio (SNR) of about 14. Figure 7 (b) shows the relationship between diameter of the sample tungsten ball and signal amplitude measured from the samples. As shown, the signal amplitude is approximately proportional to the cubic of the sample diameter. Since the maximum peak-to-peak noise amplitude of the system was about 0.5 pT, it is demonstrated that a tungsten ball of diameter of about 15 μm can be detectable by the system with the SNR of at least 2.

![Figure 4.](image)

**Figure 4.** (a) Block diagram of magnetic contaminant detection system consisting of HTS-rf-SQUID in microscopic cryostat, electronics, magnetic and electromagnetic cases, belt conveyers, permanent magnet, filters, and PC.

(b) HTS-rf-SQUID covered with HTS films installed in microscopic cryostat.
5. Conclusion
The YBCO HTS-rf-SQUID was designed and fabricated on the bicrystal STO substrate. By covering the GB and/or the slit of the SQUID with the films on the normal STO substrates in the flip-chip configuration, the effective area and the 1/f noise profile of the SQUID were successfully improved. With the two films on both the GB and the slit, the SQUID had not only the largest effective area of 0.93 mm², but also relatively large noise of 1 – 2 pT/Hz½ at 1 Hz. By installing the covered SQUID in the magnetic contaminant detection system, it is successfully demonstrated that the system has a potential to detect a tungsten ball of 15 μm in diameter with the SNR of at least 2.

Acknowledgements
This work is partially supported by “The Knowledge Hub” of Aichi, The Priority Research Project.
References

[1] Takemoto M, Akai T, Kitamura Y, Hatsukade Y and Tanaka S 2011 IEEE Trans. Appl. Supercond. 21 522
[2] Chen J-C, Chen K-L, Yang H-C, Wu C-H and Horng H-E 2007 Appl. Phys. Lett. 90 153504
[3] J. Clarke and A. I. Braginski (Eds.) 2004 The SQUID Handbook Vol. I (Wiley, Weinheim,) p 14
[4] Ketchen M B 1991 IEEE Trans. Magn. 27 2916
[5] http://www.jsquid.com/index.htm/
[6] Zhang Y, Schubert J, Wolters N, Banzet M, Zander W and Krause H-J 2002 Physica C 372-376 282
[7] Zhang Y, Yi H R, Schubert J, Zander W, Krause H-J, Bousack H and Braginski A I 1999 IEEE Trans. Appl. Supercond. 9 3396
[8] Hirano S, Oyama H, Matsuda M, Morooka T, Nakayama S and Kuriki S 2001 IEEE Trans. Appl. Supercond. 11 925
[9] Straub R, Keil S, Kleiner R and Koelle D 2001 Appl. Phys. Lett. 78 3645