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Abstract
We have developed planar nanoSQUID with nanobridge-type Josephson junctions based on the oxidation resistant and high $H_C$ MoRe alloy. The objective of the research was to reduce size of the SQUID loop with the aim being to reduce magnetic flux noise and improve the spatial resolution of the SQUID sensors. Employing RF-magnetron sputtering, electron-beam lithography, and reactive ion etching in CHF$_3$ + O$_2$ plasma using Al hard masks, we have realized nanoSQUIDs with Josephson junctions in the form of 30 – 50 nm wide nanobridges and an effective magnetic flux capture radius of ~ 95 nm. The critical temperature of the fabricated devices was $T_c = 7.9$ K. The $I(V)$-characteristics demonstrated critical current $I_0 \approx 114 \mu$A at 4.2 K and modulation period in magnetic fields of ~ 700 Oe.

Keywords: nanobridges, nanoSQUID, MoRe, Molybdenum Rhenium, scanning SQUID microscopy

(Some figures may appear in colour only in the online journal)

1. Introduction

Scanning magnetic imaging can be realized with various types of magnetic microscopy [1–5]. Among them, the SQUID-based scanning microscope has the best sensitivity, much less than the flux quantum $\sim 2 \times 10^{-15}$ Wb [3, 6] and a typical experimental spectral density of flux noise less than $1 \times 10^{-6} \Phi_0/\sqrt{\text{Hz}}$, whereas theoretical flux noise $< 1 \times 10^{-15} \Phi_0/\sqrt{\text{Hz}}$ can be achieved in hybrid superconducting structures at temperatures below 1 K [7]. However, improvement of spatial resolution is still a challenging task. The apparent solution of this limitation seems to be a decrease in the geometrical dimensions of the SQUID [8–12]. It is the essential reason why SQUIDs of nanoscale dimensions attract the interest and efforts of numerous groups worldwide. The most impressive success in magnetic and thermal nano-imaging was demonstrated recently using SQUID-on-tip (SOT) systems [8, 13, 14]. Nevertheless, the fabrication of SOT is a too delicate task. The planar geometry of nanoSQUIDs gives a good balance between a reliable nano-fabrication, high sensitivity and spatial resolution [6, 11, 15–18].

Direct-current SQUIDs (dc SQUIDs) consists of a loop of superconductor with two Josephson junctions [19]. Considering Josephson junctions (JJ) shunted with capacitance $C$ and normal state resistance $R$ of the JJ [20, 21], using the resistively and capacitively shunted junction (RCSJ) model, one can yield the resulting current $I$ through JJ as

$$I = I_0 \sin \varphi + \frac{V(t)}{R} + C \frac{dV(t)}{dt},$$

(1)
where \( I_0 \), \( \varphi \), \( V \) are the critical current, phase difference of wave functions of superconductors, and potential on the junction, correspondingly. Introducing a normalized time \( \tau \equiv (2eI_0R/\Phi_0) \) and after the substitution \( \beta_c = 2\pi I_0 CR^2/\Phi_0 \) (\( \Phi_0 = \pi h/e \) is the magnetic flux quantum), from the second Josephson equation we get

\[
\frac{d\varphi}{dt} = \frac{\beta_c}{2} \frac{d^2\varphi}{dt^2} + \frac{\Phi_0}{\Phi_0} \sin \varphi
\]

(2)

In magnetic field \( B \) the phase differences at the first and second JJ can be written as [22]

\[
\varphi_1 - \varphi_2 = 2\pi \frac{\Phi}{\Phi_0} = 2\pi \frac{\Phi_{ext} + LI_p}{\Phi_0}.
\]

(3)

In the last equation, \( \Phi \) is the total flux through the SQUID loop, \( \Phi_{ext} = BA_{eff} \) is the flux through an effective area \( A_{eff} \) and the circulating persistent current \( (I_p) \) contribution is \( LI_p \). The SQUID inductance \( L \) has both kinetic and geometric contributions. Along with the Stewart-McCumber parameter \( \beta_c \), the screening parameter \( \beta_L = 2LI_p/\Phi_0 \) is the next most significant coefficient for SQUIDs [9, 23]. If the value of \( \beta_L \ll 1 \), then after solving equations (3) and (2) jointly, the critical current can be represented as

\[
I_c = \sqrt{(I_1 - I_2)^2 + 4I_1I_2\cos^2\left(\frac{\pi\Phi}{\Phi_0}\right)}.
\]

(4)

Otherwise, the modulation of critical current could be expressed as [23]

\[
\Delta I_c/I_c \approx 1/(1 + \beta_L).
\]

(5)

NanoSQUIDs were proposed as a low noise alternative to traditional SQUIDs with superconductor-insulator-superconductor (SIS) JJs to reach a single electron spin flip sensitivity. The single spin flip is equivalent to one Bohr magneton detection, \( \mu_B \mathcal{H} = e\hbar/2m = 9.27 \times 10^{-24} \text{ J/T} \), where \( e \) is the elementary charge, \( h \) is the reduced Planck constant and \( m \) is the electron mass [9]. NanoSQUIDs have advantages over traditional SQUIDs due to lower inductance, smaller loop diameter as well as more straightforward fabrication in a single patterning step [9]. Optimizations of low-noise nanoSQUIDs are multiparametric, which combine technology of nanofabrication, materials used, geometrical design and so on, all these parameters being aimed to increase sensitivity, decrease noise level, improve spatial resolution of magnetic field detection, widen temperature range (10 mK - 10 K) and realize high magnetic field operation (\( 1 - 5 \) T). For nanoSQUID fabrication, Dayem nano-bridges [24] (DB) are commonly used [16, 25, 26] as JJs. A DB is a superconducting weak link formed with a small neck between two massive superconductors of the same material. It is essential that the dimension of this neck should be compatible with the superconductor coherence length [11]. DB seemed to be very promising for the facile and straightforward design of planar SQUIDs for high resolution magnetic force microscopy.

From a material point of view, Nb [27], Al [28], NbN [29, 30], MoRe [13] are typically used for temperatures \( T > 1 \) K.

Compared to the above mentioned superconductors, thermal noise is reduced by the choice of superconducting material with lower critical temperatures \( T_c \) (Ti, MoAu bilayers, AlMn alloys). It should be noted that 1/f noise in superconducting nanostructures is mainly determined by the two-level systems in imperfect oxides on the surface of active to oxidation metals (Al, Nb, Ta, Ti) or defects on metal-substrate interfaces [31–33]. Therefore, very important point of low-noise nanoSQUID realization is the usage of resistant to oxidation superconductors, such as NbN, TiN, NbTiN, MoRe [34, 35] and careful substrate preparation before superconductor deposition. Positive qualities of very thin MoRe films (2 – 10 nm) are both preservation of superconducting properties [36] and the robustness of superconductivity to nanofabrication techniques [37], which is favorable for fabrication of 10 – 30 nm wide superconducting nano-contacts. MoRe has already been successfully used for SOTs with high performance [13], motivating us to use it in this work. We decided to investigate a Molybdenum-Rhenium (MoRe 60-40) alloy as a material for planar nanoSQUID preparation. MoRe, for 1 - 3 alloys demonstrate superconducting transition temperatures 8 – 13 K and upper critical field higher 3 T, depending on the alloying ratio. MoRe is a type-II superconductor for which the magnetic and transport properties have been studied since the 1960’s [38–41]. The employment of MoRe 1 - 3 alloy for s-wave superconductors in JJs was demonstrated recently [42]. MoRe thin films are relatively easily achieved by magnetron sputtering, slightly susceptible to oxidation at ambient conditions, and suitable for electron beam lithography.

### 2. Sample preparation

Recently, some sophisticated methods of nanoSQUID preparation were reported [12, 43–46]. However, MoRe planar nanoSQUIDs with ring diameter ~ 100 nm, fabricated with
e-beam lithography technique, seems to us most feasible, handy, and reproducible.

In this paper, we use high-quality magnetron deposited MoRe thin films [36] in combination with the development of nanofabrication using aluminum hard masks and plasma etching. The fabrication technology is schematically shown in figure 1. The Si/SiO$_2$ 270 nm thickness substrate was purified in n-methylpyrrolidone (NMP) at a temperature of 80°C for 20 minutes in an ultrasonic bath and then rinse thoroughly with water. Superconducting MoRe was deposited on the prepared substrate in a magnetron sputtering unit with a preliminary vacuum of 10$^{-6}$ mbar. Before sputtering, the substrate was pre-treated in an argon plasma. Then 21 nm Mo(60)–Re(40) was spun. For masking, a positive electronic resist AR-P 6 400.04 (CSAR 62) was deposited and spin-coated at 4500 rpm for 1 minute. After that, the sample was baked on a hot plate at 160°C for 5 minutes. The resulting resist thickness was 80 nm.

Electron-beam lithography was carried out on the Crestec Cable 9000 machine with a minimum electron beam diameter of 10 nm (exposure 140 μC cm$^{-2}$, at 1 μs). As the lithography process was completed, the resist was developed in AR 600-546 for 1 minute, and the samples were cleaned in isopropyl alcohol. It was experimentally established that the smallest reproducible width of the bridges was about 30 nm.

To create a mask for the etching technology, an aluminum layer of 20 nm thickness was deposited using the MEB 550S Plassys electron-beam evaporator with a rate of 0.5 nm/s in a vacuum of 10$^{-6}$ mbar. After Al deposition, the sample was placed in NMP at 80°C for 1 hour for next lift-off process. For removing MoRe regions unprotected by the Al-mask, the sample was etched in CHF$_3$ + O$_2$ plasma for 2 minutes on a Corial 200i setup. Finally, the Al hard mask was removed in a 1% KOH water solution. The resulting nanoSQUID was formed as a ring including two thin bridges connecting two superconducting leads. The SEM images (Jeol 7001f) of our samples are demonstrated in figure 2.

Figure 2. SEM image of the MoRe planar nanoSQUID. a) An overall view of samples with marked current and potential leads for quasi-4-probe measurements. b) The close-up image of Dayem bridges with widths 31 and 48 nm. Green ellipse presents an effective magnetic flux capture area $A_{eff}$. An equivalent circuit of the SQUID is shown in the insert.

3. Results and discussion

The nanoSQUID samples with DBs were prepared as described above. We took two factors for the sample geometry into consideration: wide leads reduce kinetic inductance, and the whole system is suitable for cantilever mounting. As a result, MoRe nanoSQUIDs with two constrictions of approximately 30–50 nm typical width were fabricated. The characterization of samples was conducted in a liquid helium cryostat in magnetic fields up to 1.4 T with a standard quasi-4-probe measurement technique.

Figure 3 demonstrates the current-voltage curve of the sample at 4.2 K. Since the electrical capacitance of DB is a negligibly small value, the McCumber parameter $\beta_c$ is a negligible quantity too. That means the hysteresis in current-voltage (I-V) characteristics would be suppressed. However, SQUIDs with DB may exhibit hysteretic behavior [22]. The explanation is that when the bridge goes into normal state, the released heat warms the whole SQUID ring and strongly reducing the re-trapping current. This causes the strong hysteresis of the I-V curve, which is easily seen in figure 3. This hysteresis is an obstacle for the implementation of this type of SQUID for scanning magnetic imaging, and one of the possible solutions to this problem is for normal metal deposition as the resistive shunt [16]. For the sample with bridges of 31 and 48 nm, presented in figure 2, the critical current $I_0$ was 114 ± 2 μA and the re-trapping current $I_r = 26 ± 2$ μA (see figure 3). Therefore, the critical currents of each JJ are $I_{11} = 45$ and $I_{12} = 69$ μA, correspondingly.

The hysteresis might also be suppressed by temperature increasing. The critical temperature of our JJ is $T_c = 7.9 ± 0.05$ K (see inset in figure 3). The set of I-V curves at various temperatures is presented in figure 4. Our data demonstrate the narrow region 5.95 – 7.9 K, where the JJs are non-hysteretic.

The measured dependence of critical current $I_c$ upon the external magnetic field $B$ (figure 5) allows to evaluate of the
properties of weak links. An estimated critical current modulation depth is given by equation 5. Putting $\Delta I_c = 15$ and $I_c = 114 \mu A$, we get $\beta I_c \approx 6.7$ and, consequently, the inductance $L \simeq 117$ pH. The inductance $L$ has two terms: geometrical inductance $L_g$ and kinetic inductance $L_k$ [47]. The inductance $L_g$ refers to the induced flux in the SQUID loop, and $L_k$ is due to the kinetic energy of Cooper pairs. The last value could be neglected, provided the width and thickness of the SQUID is much smaller than the London penetration depth $\lambda_L$. The simple evaluation one can apply: for a ring with a radius $R$ and width $d \ L_g \sim \mu_0 R \ L_k = \frac{4 \pi \lambda_L^2}{d}$, where $\mu_0$ is the vacuum permeability. For our samples, the geometry inductance is in order of 0.1 pH, therefore the inductance $L$ is determined by the kinetic component. It gives us an approximate estimate of $\lambda_L \sim 500$ nm.

The NanoSQUID is asymmetric, showing different slopes of critical current $dI_c/dB$ on opposite sides of each maxima. The period of critical current oscillations with the magnetic field (see figure 5) is $B \approx 700$ Oe, which corresponds to the effective magnetic flux capture area $A_{eff} = \Phi_0 / B \approx (29 \pm 3) \times 10^{-3} \ \mu m^2$. Since the bridges width $d \ll \lambda_L$, the magnetic field is not screened entirely and currents may go over the full surface. The colored ellipse in figure 2 with semi-axes 66 and 139 nm represents the effective surface area of the SQUID.

4. Conclusion and further work

With this work, we demonstrated the possibility to fabricate MoRe planar nanoSQUIDs for prospective implementation for thermal and magnetic imaging, prepared with an electron-beam lithography technique. Moreover, MoRe alloy was employed for the first time for this type of nanoSQUID. The obtained samples have the superconducting transition temperature $T_c = 7.9$ K, the critical current at $T = 4.2$ K is $I_c = 115 \mu A$, the re-trapping current is $I_s = 24 \mu A$. Current-voltage characteristics show the hysteretic behavior is up to almost 5.8 K that indicates a multivalued non-sinusoidal current-phase dependence. The estimated magnetic penetration depth $\lambda_L \sim 500$ nm and effective magnetic flux capture...
area $A_{eff} \simeq 30 \times 10^{-3}$ $\mu m^2$. Also, we have performed magnetometry measurements on the nanoSQUIDs at $T = 4.2$ K. The modulation depth was $\Delta I_c/I_c \simeq 13\%$ with magnetic field period $B \simeq 700$ Oe. Our findings are in good agreement with the results reported by other groups.

We consider the obtained experimental results and evaluations as satisfactory, but further development of nanoSQUIDs based on thin MoRe films is required. However, these estimations might be incorrect for hysteretic current-voltage characteristics. We associate this behavior with the fact that the dimensions of the bridges is much larger than the coherence length and, accordingly, the current-phase dependence is very distinct from the sine function dependence. In our next work, we intend to measure noise and spin spectral densities, improve the technology to reduce capture area, implement metal shunts, and optimize the nanoSQUID parameters for magnetic imaging.

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The authors declare no conflicts of interest.

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