The planar MoRe-based dc nanoSQUID

A.G. Shishkin, O.V. Skryabina, I. Rojkov, S. Dizhur, M. Faley, A. Golubov, V.S. Stolyarov

Abstract—In the present paper, we propose the planar nano-SQUID with Dayem bridges, based on s-wave MoRe alloy. It is well known, a decrease of SQUID ring radius leads to a reduction of signal noise and, therefore, to an increase in the spatial resolution and sensitivity of the device. Employing electron-beam lithography and RF-magnetron sputtering, we have developed nano-SQUIDs with $30\ldots 50\,$ nm width of bridges and effective magnetic flux capture area $\sim 30\cdot 10^{-3}\,$ μm^2 . The critical temperature of fabricated devices was $T_c=8.1\,$ K, and magnetotransport characteristics demonstrated sufficiently large critical current $I_0\simeq 114\,$ μA . The oscillations of the critical current with magnetic fields had a period $\sim 700\,$ Oe.

Index Terms—superconductivity, SQUID, .

I. INTRODUCTION

HE problem of scanning magnetic imaging can be fulfilled with various types of microscopy [1], [2], [3], [4], [5]. Among them, the scanning microscope based on SOUID (superconducting quantum interference devices) has the best sensitivity down to flux quantum $\sim 2 \times 10^{-15}$ Wb [3], [6]. However, the spatial resolution still challenging task. The apparent solution of this limitation seems to be a decrease in geometrical dimensions of SQUID [7], [8], [9], [10], [11]. This is the main reason why SQUIDs of nanoscale dimensions attract the interest and efforts of numerous groups worldwide. The most impressive success in magnetic and thermal nanoimaging was demonstrated recently on SQUID-on-tip (SOT) systems [12], [7], [13]. However, the fabrication of SOT still complicated task. The planar geometry of nanoSQUIDs gives a good balance between a comparative facile manufacturing, high sensitivity, and spatial resolution [14], [6], [15], [16], [17], [10].

It is well known, a dc SQUID is a couple of Josephson junctions (JJ) in parallel [18]. Considering JJ as a current-phase dependent source J, shunted with capacitance C and load R [19], [20], from the first Josephson's equation one can yield the resulting current I through JJ as

$$I = I_0 \sin \varphi + \frac{V(t)}{R} + C \frac{dV(t)}{dt},\tag{1}$$

where I_0 , φ , V are the critical current, phase difference of wave functions of superconductors, and potential on the junction, correspondingly. Introducing a normalized time $\tau \equiv$

M. Shell was with the Department of Electrical and Computer Engineering, Georgia Institute of Technology, Atlanta, GA, 30332 USA e-mail: (see http://www.michaelshell.org/contact.html).

J. Doe and J. Doe are with Anonymous University.

Moscow Institute of Physics and Technology (National Research University), Moscow, Russia

Institute of Solid-State Physics, Russian Academy of Sciences, Chernogolovka, Russia

Dukhov Research Institute of Automatics (VNIIA), Moscow, Russia Kotelnikov Institute of Radioengineering and Electronics of Russian Academy of Sciences, Moscow, Russia. $(2eI_0R/\Phi_0)t$ and after the substitution $\beta_C=2\pi I_0CR^2/\Phi_0$ $(\Phi_0=\pi\hbar c/e)$ is the magnetic flux quantum), from the second Josephson's equation we get

$$\frac{I}{I_0} = \sin \varphi + \frac{d\varphi}{d\tau} + \beta_C \frac{d^2 \varphi}{d\tau^2} \tag{2}$$

In magnetic field B the phase differences at first and second JJ can be written as [21]

$$\varphi_1 - \varphi_2 = 2\pi \frac{\Phi}{\Phi_0} = 2\pi \frac{\Phi_{ext} + LJ}{\Phi_0}.$$
 (3)

In the last equation Φ is is a total flux through the SQUID loop, $\Phi_{ext} = BA_{eff}$ is a flux through an effective area A_{eff} , and circulating current contribution is LJ. The SQUID inductance L has both kinetic and geometric contribution. Along with Stewart-McCumber parameter β_C , the screening parameter $\beta_L = 2LI_0/\Phi_0$ is next most important coefficient for SQUIDs [8], [22]. If the value of $\beta_L \ll 1$, solving the equations 3 and 2 jointly, the critical current can be represented

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

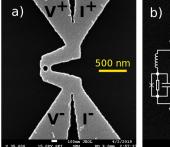
Otherwise, the modulation of critical current could be expressed as [22]

$$\Delta I_c/I_c \simeq 1/(1+\beta_L). \tag{5}$$

For fabrication JJ, Dayem bridges [23] (DB) are commonly used [24], [25], [15]. DB is a superconducting weak link formed with a small neck between two massive superconductors of the same material. It is essential, the dimension of this neck should be compatible with superconductor coherence length [10]. DB seemed to be very promising for the simple and facile design of planar SQUIDs for magnetometry microscopy. The conventional materials for DB are Ti, Ni, Al, Pb, Nb. We decided to investigate an alloy of Molybdenum-Rhenium (MoRe 60-40) alloy as a material for planar nano-SQUID preparation. MoRe is a type-II superconductor which magnetic and transport properties have been studied since 1960's [26], [27], [28], [29]. Mo_xRe_{1-x} alloys demonstrate superconducting transition temperatures 8...13 K and upper critical field greater 3 T, depending on the alloying ratio. MoRe thin films are relatively easy achieved by magnetron sputtering, slightly susceptible to oxidation at ambient conditions, and suitable for electron beam lithography.

II. SAMPLE PREPARATION

Recently, some sophisticated methods of nano-SQUID preparation were reported [30], [31], [32], [33], [11]. However, MoRe planar nano-SQUID with ring diameter $\sim 100\,$ nm, fabricated with e-beam lithography technique, seems to us most feasible, handy, and reproducible.



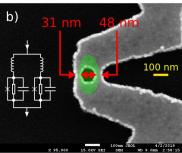


Fig. 1. SEM image of the MoRe planar nano-SQUID. a) An overall view of samples with marked current and potential leads for quasi-4-probe measurements. b) The close-up image of Dayem bridges with widths 31 and 48 nm. Green ellipse presents an effective magnetic flux capture area A_{eff} . An equivalent circuit of the SQUID in in the insert.

To fabricate such samples, we used magnetron sputtering technology [34] with etching with an aluminum mask. The $\mathrm{Si/SiO_2}$ 270 nm thickness substrate was purified in nmethylpyrrolidone (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 $3\cdot 10^{-8}$ mbar. Before sputtering, the substrate was pre-treated in an argon plasma. Then 21 nm $\mathrm{Mo}(60) - \mathrm{Re}(40)$ was sprayed. For masking, a positive electronic resist AR-P 6400.04 (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 width was 80 nm.

Electron lithography was carried out on *Crestec Cable 9000* machine with a minimum electron beam diameter of 10 nm (exposition 140 μ c/cm², at 1 μ s). 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 could be achieved with a resolution of 30 nm

To create a mask for the etching technology, an aluminum layer of 20 nm thickness was sprayed using the *MEB 550S Plassys* electron-beam evaporator with the rate 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 Al-mask, the sample was etched in CHF₃ + O_2 plasma for 2 minutes on a *Corial 200i* setup. For subsequent removal of Al, the substrate was placed in a 1% KOH solution.

The resulting nano-SQUID was formed as a ring included two thin bridges connecting two superconducting leads. The SEM images (*Jeol 7001f*) of our samples are demonstrated in Fig.1).

III. RESULTS AND DISCUSSION

The nano-SQUID samples with DBs were prepared as described above. We took two reasons for the sample geometry into consideration: wide leads reduce kinetic inductance, and the whole system is suitable for cantilever mounting. As a result, MoRe nano-SQUIDs with two constrictions of

approximately 30...50 nm typical width were fabricated. The characterization of samples was conducted in liquid helium cryostat in magnetic fields up to 1.4 T by standard quasi-4-probe measurement technique.

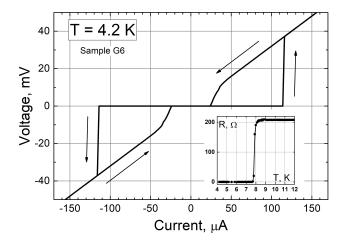


Fig. 2. Current-Voltage characteristics of the sample with two Dayem bridges. The critical current is 114 $\,\mu\mathrm{A}$, re-trapping current is 26 $\,\mu\mathrm{A}$. The temperature dependence of the sample resistance R(T) measured at 5 $\,\mu\mathrm{A}$, is in the insert. The critical temperature $T_c=8.1\,$ K.

Figure 2 demonstrates the current-voltage curve of the sample at 4.2 K. Since the capacity of DB is negligibly small value, McCumber parameter β_C is a negligible quantity also. That means the hysteresis in current-voltage (I-V) characteristics would be suppressed. However, SQUIDs with DB may exhibit hysteretic behavior [21]. The explanation is the following: when the bridge goes into normal state, the released heat warms the whole SQUID ring and the critical current decreases. It causes the strong hysteresis of I-V curve, which well seen in Fig. 2. This hysteresis is an obstacle for the implementation this type of SQUID for scanning magnetic imaging, and one of the possible solutions to this problem is in normal metal deposition as resistive shunt [15]. For the sample with bridges of 31 and 48 nm, presented in Fig. 1, the critical current I_0 was 114 ± 2 μA and re-trapping current $I_r = 26 \pm 2 \, \mu \text{A}$ (see Fig. 2). Therefore, the critical currents of each JJ are $I_{c1}=45$ and $I_{c2}=69~\mu\text{A}$, correspondingly.

The hysteresis might be suppressed by temperature increasing too. The critical temperature of our JJ is $T_c=8.1\pm0.05$ K (see insert in Fig 2). The set of I-V curves at various temperatures is presented in Fig. 3. Our data demonstrate the narrow region $5.95\ldots6.14$ K, where superconductivity still exists, but hysteresis vanishes.

The measured dependence of critical current I_c upon external magnetic field B (Fig 4) allows evaluating the properties of weak links. An estimated critical current modulation depth is given by Eq. 5. Putting $\Delta I_c = 15$ and $I_c = 114~\mu A$, we get $\beta_L \simeq 6.7$ and, consequently, the inductance $L \simeq 117~{\rm pH}$. The inductance L has two terms: geometrical inductance L_g and kinetic inductance L_k [35]. The inductance L_g refers to induced flux in the SQUID circle, and the L_k is due to kinetic energy of Cooper pair. Last value could be neglected, provided

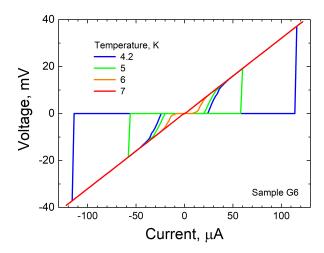


Fig. 3. The family of current-voltage curves at various temperatures.

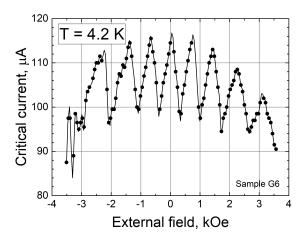


Fig. 4. The oscillation of critical current I_C in external magnetic field B at 4.2 K. The period of oscillation $T_B\simeq 700\pm 60\,$ Oe

the width and thickness of the SQUID loop much large than London penetration depth λ_L . The simple evaluation one can apply: for a ring with a radius R and width d $L_g \sim \mu_0 R$ and $L_k = 4\pi\lambda_L^2/d$, where μ_0 is vacuum permeability. For our samples, the geometry inductance is in order of 0.1 pH, therefore the inductance L is determined by kinetic component. It gives us an approximate estimate of $\lambda_L \sim 500\,$ nm.

The period of critical current oscillations with the magnetic field (Fig 4 is $T_B \simeq 700\,$ Oe, which corresponds the effective magnetic flux capture area $A_{eff} = \Phi_0/T_B \simeq (29\pm3)\times 10^{-3}\,$ $\mu\mathrm{m}^2$. 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 Fig 1 with semi-axes 66 and 139 nm represents the effective surface area of the SQUID.

We have estimated the noise characteristics of our SQUID. The SQUID resistance (from IV-diagram) is $\simeq 0.3$ Ohm, hence each JJ has $R_n \simeq 0.6$ Ohm. From numerical evaluation in [22], we can get

$$\frac{S_{\Phi}}{2L} \simeq \frac{S_V}{(\partial V/\partial \Phi)^2 2L_k} \simeq \frac{16k_B T R_n}{(\partial V/\partial \Phi)^2 2L_k},\tag{6}$$

where S_{Φ} is flux noise, and k_B is Boltzmann constant. The energy resolution E is $483\hbar$ approximately; flux noise $S_{\Phi} \simeq \sqrt{E \cdot 2 \cdot L}/\Phi_0$ and spin noise $S_n \simeq S_{\Phi} r/r_e$ (r_e is classical electron radius and considering effective area A_{eff} as a circle, r is a radius of this area), will give us $1.7 \cdot 10^{-6}$ $\Phi_0/\sqrt{\rm Hz}$ and $103~\mu V/\sqrt{\rm Hz}$, respectively.

IV. CONCLUSION AND FURTHER WORK

With this work, we demonstrated the possibility to fabricate MoRe planar nano-SQUIDs for possible implementation for thermal and magnetic imagine, prepared with electron lithography technique. Moreover, MoRe alloy was employed for the first time for this type of nano-SQUIDs. The obtained samples have superconducting transition temperature $T_c = 8.1$ K, the critical current at T=4.2 K $I_C=115$ μA , the retrapping current is $I_r = 24 \mu A$ Current-voltage characteristics show the hysteretic behavior up to almost 5.8 K that indicates overheating the structure. The estimated magnetic penetration depth $\lambda_L \sim 500\,$ nm and effective magnetic flux capture area $A_{eff} \simeq 30 \cdot 10^{-3} \ \mu \text{m}^2$. Also, we have performed magnetometry measurements on the nano-SQUIDs at $T=4.2\,$ K. The modulation depth was $\Delta I_c/I_C \simeq 13\%$ with modulation period $T_B \simeq 700\,$ Oe. Our findings are in good agreement with the results reported by other groups.

We consider the obtained experimental results and evaluations as very satisfactory for nano-SQUIDs. 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. In our next work, we intend to measure noise and spin spectral densities, improve the technology to reduce capture area, implement metal shunt, and optimize nano-SQUID parameters for magnetic imagine.

The authors declare no conflicts of interest.

ACKNOWLEDGMENT

The authors would like to thank...

This work was supported in parts by Russian Foundation for Basic Research (RFBR) grant No

REFERENCES

- O. Kazakova, R. Puttock, C. Barton, H. Corte-León, M. Jaafar, V. Neu, and A. Asenjo, "Frontiers of magnetic force microscopy," *Journal of Applied Physics*, vol. 125, no. 6, p. 060901, 2019.
- [2] J. R. Kirtley, "Fundamental studies of superconductors using scanning magnetic imaging," *Reports on Progress in Physics*, vol. 73, no. 12, p. 126501, nov 2010.
- [3] J. R. Kirtley and J. P. Wikswo, "Scanning squid microscopy," Annual Review of Materials Science, vol. 29, no. 1, pp. 117–148, 1999.
- [4] E. Polturak, "Beyond the horizon: Magneto-optical imaging studies of the kibble-zurek scenario in superconductors," *Journal of Low Temperature Physics*, Jul 2019.
- [5] M. Buchner, K. Höfler, B. Henne, V. Ney, and A. Ney, "Tutorial: Basic principles, limits of detection, and pitfalls of highly sensitive squid magnetometry for nanomagnetism and spintronics," *Journal of Applied Physics*, vol. 124, no. 16, p. 161101, 2018.

- [6] T. Golod, O. Kapran, and V. Krasnov, "Planar superconductor-ferromagnet-superconductor josephson junctions as scanning-probe sensors," *Phys. Rev. Applied*, vol. 11, p. 014062, Jan 2019.
- [7] D. Vasyukov, Y. Anahory, L. Embon, D. Halbertal, J. Cuppens, L. Neeman, 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 Nanotechnology*, no. 9, pp. 639–644, sep.
- [8] C. Granata and A. Vettoliere, "Nano Superconducting Quantum Interference device: A powerful tool for nanoscale investigations," *Physics Reports*, pp. 1–69, feb.
- [9] D. Hazra, "Nanobridge superconducting quantum interference devices: Beyond the josephson limit," *Phys. Rev. B*, vol. 99, p. 144505, Apr 2019. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB. 99.144505
- [10] W. Wernsdorfer, "From micro-to nano-SQUIDs: Applications to nano-magnetism," Superconductor Science and Technology, no. 6, p. 064013, inn.
- [11] C. P. Foley and H. Hilgenkamp, "Why NanoSQUIDs are important: an introduction to the focus issue," *Superconductor Science and Technol*ogy, no. 6, p. 064001, jun.
- [12] K. Bagani, J. Sarkar, A. Uri, M. L. Rappaport, M. E. Huber, E. Zeldov, and Y. Myasoedov.
- [13] D. Halbertal, M. Ben Shalom, A. Uri, K. Bagani, A. Y. Meltzer, I. Marcus, Y. Myasoedov, J. Birkbeck, L. S. Levitov, A. K. Geim, and E. Zeldov, "Imaging resonant dissipation from individual atomic defects in graphene," vol. 358, no. 6368, pp. 1303–1306, 2017.
- [14] M. Ketchen, D. Awschalom, W. Gallagher, A. Kleinsasser, R. Sandstrom, J. Rozen, and B. Bumble, "Design, fabrication, and performance of integrated miniature SQUID susceptometers," *IEEE Transactions on Magnetics*, no. 2, pp. 1212–1215, mar.
- [15] S. K. H. Lam and D. L. Tilbrook, "Development of a niobium nanosuperconducting quantum interference device for the detection of small spin populations," *Applied Physics Letters*, vol. 82, no. 7, pp. 1078– 1080, 2003.
- [16] R. Kleiner, D. Koelle, F. Ludwig, and J. Clarke, "Superconducting quantum interference devices: State of the art and applications," *Proceedings of the IEEE*, no. 10, pp. 1534–1548, oct.
- [17] R. L. Fagaly, "Superconducting quantum interference device instruments and applications," *Review of Scientific Instruments*, no. 10, p. 101101, oct.
- [18] P. W. Anderson and A. H. Dayem, "Radio-frequency effects in superconducting thin film bridges," *Phys. Rev. Lett.*, vol. 13, pp. 195–197, Aug 1964.
- [19] W. C. Stewart, "Currentvoltage characteristics of josephson junctions," Applied Physics Letters, vol. 12, no. 8, pp. 277–280, 1968.
- [20] D. E. McCumber, "Effect of ac impedance on dc voltagecurrent characteristics of superconductor weaklink junctions," *Journal of Applied Physics*, vol. 39, no. 7, pp. 3113–3118, 1968.
- [21] J. Clarke and A. I. Braginski, Weinheim.
- [22] C. D. Tesche and J. Clarke, "dc squid: Noise and optimization," *Journal of Low Temperature Physics*, vol. 29, no. 3, pp. 301–331, Nov 1977.
- [23] P. W. Anderson and A. H. Dayem, "Radio-frequency effects in superconducting thin film bridges," *Phys. Rev. Lett.*, vol. 13, pp. 195–197, Aug 1964.
- [24] W. Keijers, X. D. A. Baumans, R. Panghotra, J. Lombardo, V. S. Zharinov, R. B. G. Kramer, A. V. Silhanek, and J. Van de Vondel, "Nano-squids with controllable weak links created via current-induced atom migration," *Nanoscale*, vol. 10, pp. 21475–21482, 2018.
- [25] R. F. Voss, R. B. Laibowitz, and A. N. Broers, "Niobium nanobridge dc squid," *Applied Physics Letters*, vol. 37, no. 7, pp. 656–658, 1980.
- [26] E. Lerner, J. G. Daunt, and E. Maxwell, "Magnetic properties of superconducting mo-re alloys," *Phys. Rev.*, vol. 153, pp. 487–492, Jan 1967
- [27] J. Gavaler, M. Janocko, and C. Jones, "A15 structure more superconductor," *Applied Physics Letters*, vol. 21, no. 4, pp. 179–180, 1972.
- [28] J. Talvacchio, M. A. Janocko, and J. Greggi, "Properties of evaporated mo-re thin-film superconductors," *Journal of Low Temperature Physics*, vol. 64, no. 5, pp. 395–408, Sep 1986.
- [29] S. Sundar, L. S. Sharath Chandra, V. K. Sharma, M. K. Chattopadhyay, and S. B. Roy, "Electrical transport and magnetic properties of superconducting mo52re48 alloy," *AIP Conference Proceedings*, vol. 1512, no. 1, pp. 1092–1093, 2013.
- [30] T. Godfrey, J. C. Gallop, D. C. Cox, E. J. Romans, J. Chen, and L. Hao, "Investigation of dayem bridge nanosquids made by xe focused ion beam," *IEEE Transactions on Applied Superconductivity*, vol. 28, no. 7, pp. 1–5, Oct 2018.

- [31] A. Finkler, Y. Segev, Y. Myasoedov, M. L. Rappaport, L. Neeman, D. Vasyukov, E. Zeldov, M. E. Huber, J. Martin, and A. Yacoby, "Selfaligned nanoscale squid on a tip," *Nano Letters*, vol. 10, no. 3, pp. 1046–1049, 2010, pMID: 20131810.
- [32]
- [33] J. Gallop and L. Hao, "Nanoscale superconducting quantum interference devices add another dimension," ACS Nano, vol. 10, no. 9, pp. 8128– 8132, 2016, pMID: 27579572.
- [34] V. A. Seleznev, M. A. Tarkhov, B. M. Voronov, I. I. Milostnaya, V. Y. Lyakhno, A. S. Garbuz, M. Y. Mikhailov, O. M. Zhigalina, and G. N. Gol'tsman, "Deposition and characterization of few-nanometers-thick superconducting MoRe films," *Superconductor Science and Technology*, no. 11, p. 115006, nov.
- [35] M. José Martínez-Pérez and D. Koelle, "NanoSQUIDs: Basics & recent advances," *Physical Sciences Reviews*, no. 8, aug.

Michael Shell Biography text here.

PLACE PHOTO HERE

John Doe Biography text here.

Jane Doe Biography text here.