Quick and reliable technology for fabrication of stand-alone BSCCO mesas

E A Vopilkin1, A V Chiginev1,2,3, L S Revin1,2,3, A N Tropanova1,3, I Yu Shuleshova1, A I Okhapkin1,3, A D Shovkun4, A B Kulakov4 and A L Pankratov1,2,3

1 Institute for Physics of Microstructures of RAS, GSP-105, Nizhny Novgorod, 603950, Russia
2 Laboratory of Cryogenic Nanoelectronics, Nizhny Novgorod State Technical University n.a. R.E. Alekseev, Nizhny Novgorod, Russia
3 Lobachevsky State University of Nizhni Novgorod, Nizhny Novgorod, Russia
4 Institute of Solid State Physics of RAS, Chernogolovka, Russia

E-mail: alp@ipmras.ru

Received 2 October 2014, revised 9 January 2015
Accepted for publication 12 January 2015
Published 6 February 2015

Abstract

The technology of wet etching allowing fabrication of stand-alone BSCCO mesa structures was proposed. The produced mesas can be made much thicker than ones usually being studied. The time required for the fabrication is much smaller in comparison with the standard method of ion milling. The process used is controllable which provides acceptable precision of mesa fabrication. The IV characteristics of the sample showing Josephson nature were obtained. The qualitative comparison with characteristics of similar structures fabricated by other groups was carried out.

Keywords: intrinsic Josephson junction, BSCCO mesa, wet etching

At the present time, the investigations of subTHz and THz radiation from Bi2Sr2CaCu2O8+x (BSCCO) mesas attract a great interest (see [1] and references therein). BSCCO is a high-Tc superconductor with high anisotropy, leading to appearance of an intrinsic Josephson effect [2]. In other words, the CuO layers of such a material appear to be coupled by Josephson coupling. Thus, BSCCO represents a stack of Josephson junctions formed on an atomic scale. Note that artificial stacks made of high-Tc materials are not available. Even junctions between low-Tc and high-Tc superconductors in c-direction are inhomogeneous, exhibiting a small critical current density and low LRp product [3]. Since the discovery of the intrinsic Josephson effect, there have been many attempts to make the Josephson junctions of the stack oscillate coherently, in order to produce strong electromagnetic radiation. However, the first success in generation and observation of the electromagnetic radiation from BSCCO mesa was attained in 2007 [4]. The observed radiation had the power of order 0.5 μW and frequency up to 0.85 THz. Since then, the radiation power has been increased to values of tens of μWs [5] and recently up to 0.6 mWs [6]. These achievements make it possible to use BSCCO mesas as oscillators in subTHz and THz range to fill in the so-called ‘THz gap’. By now, the obtained power and spectral line-width of the electromagnetic radiation from BSCCO mesas [5–7] allow using them as sources for molecular spectroscopy [8].

Traditionally, the technology of mesa fabrication is based on the method of ion milling. On the one hand, it allows to produce mesas with high precision and minimize the mechanical impact on the material. On the other hand, this technology does not allow to fabricate mesas thicker than 1–2 μm, due to the slowness of the ion milling process. At the same time, making the thicker mesas could involve more intrinsic junctions into synchronization and thus enhance the radiated power. The advantages of the stand-alone mesas are well-defined symmetric electromagnetic boundary conditions, anticipated high emission power, and more efficient cooling. Besides, better thermal conductivity of stand-alone mesas allows making thicker structures without risk of overheating.

In this paper we report on the technology of mesa fabrication, based on a wet etching. This technology allows to
obtain thicker mesas, and is still controllable to provide acceptable precision in mesa fabrication.

The research aimed for the fabrication of a single BSCCO mesa structure for use as a subTHz range generator was performed. For this purpose, a few mm lead-doped Bi$_{1.85}$Pb$_{0.35}$Sr$_2$CaCu$_2$O$_{8+x}$ flakes with $T_c \approx 90$ K were used, for details see [9]. These flakes were obtained by cleaving the piece of BSCCO single crystal, grown from the melt in gold crucible [9–11]. The flakes were mounted by the conductive glue to the silicon substrate coated with gold. We note that here and below we use acronym BSCCO for Bi$_{1.85}$Pb$_{0.35}$Sr$_2$CaCu$_2$O$_{8+x}$ as well.

For the fabrication of BSCCO mesa structures the ion milling is commonly used [4, 12, 13]. In our work in the same way the thick mesa was tried to be milled by argon ion bombardment in the Oxford Plasmalab 80 Plus facility. The milling was carried out in 20 s cycle. After twenty cycles the ion-milling facility required cooling. The depth of milling in 20 cycles was 60 nm, therefore the milling rate was about 2 nm per cycle. This method is obviously not convenient for fabrication of a mesa structure with a few microns depth since it requires too much time of the facility operation.

In our study the experiments on BSCCO wet etching were undertaken. For this purpose, the 100 × 300 μm photoresist (PR) mask of 1 μm thickness was put on the surface of flakes by photolithography (figure 1(a)). According [14] the BSCCO etching use to be performed in the phosphoric acid. We have also tried to use phosphoric acid but have found that this method is not suitable for deep etching. During etching, the insoluble precipitates are rapidly formed on the surface of BSCCO, impeding access of etchant to the surface and greatly reducing the etching rate. After that, the experiments on BSCCO etching in hydrochloric acid (its 1:8 water solution) were carried out using the same PR mask (etching time was about 50 s). The etching was monitored visually and stopped with the disappearance of most of the flakes not covered with PR. The result of the cross-cut etching with unremoved PR is shown in figure 1(b). It can be seen that the obtained stand-alone mesa strongly differs from the mask by the size. The undercut depth is about 40 μm. It can be seen that the mesa has fairly sharp edges. The etching was cross-cut not everywhere, because of the inhomogeneity of the flake thickness. On the most part of the surface the detached mesas appear, but there were also places where the etching was not cross-cut. Optical measurement of etching depth was carried out for these mesas. For this purpose, the PR mask was removed by plasma chemical etching in an oxygen plasma of Oxford Plazmalab 80 Plus facility. Etching results are shown in figure 1(c). The height of the mesa (figure 2) was measured using a Talysurf 2000 white light interferometer, and is

Figure 1. The original BSCCO sample with photomasks (a), the result of the cross-cutting etching of BSCCO single crystal (b) and the same sample after removing of the photomask (c).

Figure 2. Talysurf 2000 white light interferometer pictures of BSCCO single crystal obtained by wet etching.
roughly 7 μm. Thus it is seen that the etching is anisotropic: undercut depth is 40 μm with an etching depth of 7 μm. The anisotropy of the etching rate allows to obtain mesas with almost vertical walls, see figure 2 and the SEM image of the circular mesa (see below, figure 4).

We note that the fabrication of BSCCO mesas in dilute HCl has been discussed elsewhere [15, 16]. In these works, the authors report about formation of a glass-like BiOCl under the treatment of the acid. However, in our work only tiny amount of a glass-like substance at the mesa edges can be observed. We assume that this is due to the fact that the authors of [15, 16] use HCl with concentration 30–50 times lower than in our study. Therefore, at lower HCl concentration BiOCl remains undissolved.

Further experiments on BSCCO etching were performed with PR mask in the form of circles with various diameters. The etching in hydrochloric acid yielded stand-alone circular mesas (see figure 3(a)) as well as mesas on the pedestal (see figure 3(b)). It is seen that the undercut is deeper on circles with smaller diameter.

Next problem was removing of the PR. Using the oxygen plasma resulted to the facility contamination, so another method was used. The PR was successfully dissolved in dimethylformamide without using an ultrasonic bath, see figure 3(c). The maximum time of PR removing was about 30 min. As it turned out, a conductive glue may also be dissolved, after which a separate mesa can be moved (figure 3(d)).

This allows gluing a separate mesa with 450 μm diameter by the conductive adhesive to the gold-coated substrate at any place (figure 5(a)). After that, a second electric wire was bonded to the mesa surface by the conductive glue, see figure 5(b). This configuration allows measuring the current–voltage characteristics along the c-axis of the BSCCO crystal.

In figure 6 the measured current–voltage characteristics of one of these structures for various temperatures are shown (arrows indicate forward and backward branches). Using the analysis of the experimental curves it is possible to obtain information about some properties and quality of the resulting
structure. It can be seen that with temperature decreasing the critical current increases and reaches a value of $I_c = 2.65 \text{ mA}$ at $T = 30 \text{ K}$. Here and below the critical current value is a current level at which the direct branch exhibits a sharp jump to a higher voltage ($V = 3.6 \text{ V}$ at $30 \text{ K}$). The critical current density for BSCCO structures is in the range of $10^{18} \text{ - } 2000 \text{ A} \cdot \text{cm}^{-2}$ [17–23]. In [11] the critical current density along the $ab$-plane of crystals identical to those used in this work (by size and chemical composition) is investigated and found to be of order $J_c \approx 1000 \text{ A} \cdot \text{cm}^{-2}$. It is known that the critical current density along the $c$-axis (Josephson critical current density) in Pb doped BSCCO is 5–20 times smaller than the critical current density along the $ab$-plane [22]. Thus, we come for our structures to the following estimation $J_c \approx 50–200 \text{ A} \cdot \text{cm}^{-2}$. In this case, for the investigated structure with $S \approx 16 \times 10^{-4} \text{ cm}^2$ area, the critical current $I_c$ must be around 80–320 mA, which is much larger than the measured one. This can be explained either by deterioration of the top or (and) bottom layer of the structure (see below) or by the fact that by somewhat reason only a part of the circular mesa exhibits superconducting properties. On the other hand, we consider much larger structures than usually in the literature. In this case simple linear estimation of the critical current density can give incorrect results. For example, in [24] the mean life time of superconducting state (directly related to the critical current value) has been studied versus the system length and it has been demonstrated that this characteristics saturates at the scale of 1–2 Josephson vortex size ($\sim 10$ Josephson lengths), depending on the system geometry.

From zero bias current up to the critical current $I_c$, there is a constant (independent of temperature) additional resistance $R_{\text{add}} = 760 \Omega$. Measurements were carried out with the three-point scheme, but the contribution of the wires to the additional resistance is small (fractions of Ohm) and can be neglected. Thus, the observed non-zero resistance $R_{\text{add}}$ of the mesa, and the nonlinearity of the $IV$ curve at low bias, is most likely caused by deteriorated top or (and) bottom layers, because we did not use the golden coating of the mesa. To characterize Josephson properties of the structure we subtract the initial resistive part of the current–voltage characteristics, figure 7. In this case, we observe a small back-bending feature of the $I$–$V$ characteristic indicating the so-called ‘hot spot’ associated with the Joule heating [7, 25, 26]. This small back-bending is typical for stand-alone mesas [27], because due to better heat sink, the ‘hot spot’ in stand-alone mesas should be less pronounced.

In conclusion, the proposed technology of wet etching allows obtaining stand-alone BSCCO mesa structure thicker
than ones usually being studied, but still allows to get mesas below 1 μm. The time required for the fabrication is much smaller in comparison with the standard method of ion milling, but the process is still controllable, which provides acceptable precision of mesa fabrication. While thicker mesas potentially allow to get larger oscillation power, it can lead to overheating that will decrease the power. To decrease the overheating one can decrease the bath temperature and, moreover, the stand-alone mesas are better cooled due to the absence of the superconducting substrate which has low thermal conductivity. The proposed technology, therefore, allows to perform an experimental study of the optimal mesa thickness in a broad range. The obtained current–voltage characteristics demonstrate Josephson nature and are qualitatively comparable with characteristics obtained by other groups.

Acknowledgments

The authors thank EV Skorokhodov (IPM RAS) for making SEM images of the mesa. This work is supported by the Ministry of Education and Science of the Russian Federation (project 3.2054.2014/K), by Program no. 1 of the Presidium of RAS, by RFBR (projects 12-02-01009 and 15-02-05869) and in the framework of Increase Competitiveness Program of Lobachevsky NNSU under contract no. 02.B.49.21.0003. The facilities of the Common Research Center ‘Physics and technology of micro- and nanostructures’ of IPM RAS were used.

References

[1] Welp U, Kadowaki K and Kleiner R 2013 Nat. Photonics 7 702
[2] Kleiner R, Steinmeyer F, Kunkel G and Mueller P 1992 Phys. Rev. Lett. 68 2394
[3] Komissinski P V, Il’ichev E, Ovsyannikov G A, Kotvytskyy S A, Grajcar M, Hlubina R, Ivanov Z, Tanaka Y, Yoshida N and Kashiwaya S 2002 Europhys. Lett. 57 585
[4] Ozyuzer L et al 2007 Science 318 1291
[5] Yamaki K, Tsujimoto M, Yamamoto T, Furukawa A, Kashiwagi T, Minami H and Kadowaki K 2011 Opt. Express 19 3193
[6] Benseman T M, Gray K E, Koshelev A E, Kwok W-K, Welp U, Minami H, Kadowaki K and Yamamoto T 2013 Appl. Phys. Lett. 103 022602
[7] Li M et al 2012 Phys. Rev. B 86 060505(R)
[8] Sabakinskaya E A, Vax V L, Kinev N V, Koshelets V P and Wang H 2013 Proc. 1st Russian Microwave Conf. pp 101–5
[9] Kulakov A B, Bldikin I K, Zver’kov S A, Emel’chenko G A, Yang G and Abell J S 2002 Physica C 371 45
[10] Kulakov A B, Kosenko A V, Zver’kov S A, Emelchenko G A and Ponomhilo I 1996 Supercond. Sci. Technol. 9 859
[11] Kulakov A B et al 2001 J. Cryst. Growth 231 194
[12] Kashiwagi T et al 2012 Japan. J. Appl. Phys. 51 010113
[13] Wang H B, Wu P H and Yamashita T 2001 Appl. Phys. Lett. 78 4010
[14] Kaur D and Gupta A K 2002 J. Phys. D: Appl. Phys. 35 729
[15] Kato T, Ishida H, Suematsu H, Yasui K and Hamasaki K 2012 Cryogenics 52 398
[16] Nishikata T, Kato T, Kotaki Y, Suematsu H, Kawakami A and Yasui K 2014 Japan. J. Appl. Phys. 53 04EJ02
[17] Motzkau H, Katterwe S O, Rydh A and Krasnov V M 2013 Physica C 491 51
[18] Latyshev Yu I, Pavlenko V N, Nevelskaya J E and Monceau P 1997 J. Alloys Compd. 251 236
[19] Batista-Leyva A J, Cobas R, Estevez-Rams E, Orlando M T D, Noda C and Altschuler E 2000 Physica C 331 57
[20] Katterwe S O, Rydh A, Motzkau H, Kulakov A B and Krasnov V M 2010 Phys. Rev. B 82 024517
[21] Tsujimoto M, Yamaki K, Deguchi K, Yamamoto T, Kashiwagi T, Minami H, Tachiki M, Kadowaki K and Klemm R A 2010 Phys. Rev. Lett. 105 037005
[22] Kambara H, Kakeya I and Suzuki M 2013 Phys. Rev. B 87 214521
[23] Moelle M and Kleiner R 1999 Phys. Rev. B 59 4486
[24] Fedorov K G and Pankratov A L 2009 Phys. Rev. Lett. 103 260601
[25] Wang H B, Gu’ennon S, Yuan J, Iishi A, Arisawa S, Hatano T, Yamashita T, Koelle D and Kleiner R 2009 Phys. Rev. Lett. 102 017006
[26] Yurgens A 2011 Phys. Rev. B 83 184501
[27] Tsujimoto M et al 2012 Phys. Rev. Lett. 108 107006