Fabrication of $NdBa_2Cu_3O_{7-x}$ interface-modified ramp-edge junctions by MOCVD

Y. NAKAJIMA
Superconductivity Research Laboratory, ISTEC, 1-10-13 Shinonome, Koto-ku, Tokyo 135-0062, Japan; Nihon University, 7-24-1 Narashinodai, Funabashi-shi, Chiba 274-8501, Japan

H. KOKUBUN*, S. ADACHI‡, Y. TARUTANI, K. TANABE, T. MORISHITA
Superconductivity Research Laboratory, ISTEC, 1-10-13 Shinonome, Koto-ku, Tokyo 135-0062, Japan
E-mail: adachi@istec.or.jp

N. IWATA, H. YAMAMOTO
Nihon University, 7-24-1 Narashinodai, Funabashi-shi, Chiba 274-8501, Japan

Ramp-edge Josephson junctions with an interface-modified barrier have been widely studied for single flux quantum (SFQ) digital devices, because they have higher I_cR_n products and a smaller I_c spread within a chip than those for other types of high-T_c Josephson junctions [1–6]. The barriers of interface-modified junctions are made from an amorphous layer formed on a superconducting base electrode by ion irradiation. This amorphous layer becomes a barrier layer through appropriate annealing and counter-layer deposition processes [7]. The superconducting counter electrode has been prepared by pulsed laser deposition (PLD) and sputtering methods so far [1–6]. In these methods, rather energetic particles are deposited during the counter-layer deposition process. It has been pointed out that these energetic particles significantly contribute to the barrier re-crystallization process and also cause a large spread of junction characteristics in some cases [5, 6].

In the present work, we have studied fabrication of ramp-edge junctions with an interface-modified barrier prepared by a method in which films are prepared by deposition of non-energetic particles, that is, metalorganic chemical vapor deposition (MOCVD). Additionally, this method has some advantages such as good film coverage and large-area deposition capability. There has been no report on the fabrication of ramp-edge junctions by MOCVD. We chose $NdBa_2Cu_3O_{7-x}$(NBCO) and Sr_2AlTaO_6 (SAT) for superconducting electrodes and insulators, respectively. NBCO is considered as a promising candidate for junction devices because a rather flat film surface is readily obtained and the surface is inactive with air [8]. The preparation of NBCO films with a thickness less than several hundred nanometers by MOCVD was previously reported by Komatsu *et al*. [9] and Kumagai *et al.* [10] Here, we firstly examined preparation conditions of 300-nm-thick NBCO films for fabrication of ramp-edge junctions. SAT has been expected as a suitable intermediate insulator because of its lattice constant approximately twice of the *a*- or *b*-axis of NBCO [11] and the low dielectric constant ($\varepsilon = 23$–30) [12]. Takahashi *et al.* [13, 14] previously reported successful preparation of the SAT films with good crystallinity by MOCVD. We fabricated the NBCO and SAT films separately using two different MOCVD rigs.

NBCO films were deposited on 10×10 mm^2 $SrTiO_3$(STO) (100) substrates in a cold-wall lateral-flow reactor by MOCVD. The substrate was heated on a quartz plate that was placed onto an inconel alloy susceptor where RF power is converted to heat. The metalorganic vapors were carried by Ar gas and mixed with O_2 gas at the entrance to the reactor. Table I (a) lists the preparation conditions for NBCO films. With the aim of achieving homogeneous deposition over a larger area, dpm-H was added to the carrier gas. We expected the dpm-H to prevent $Cu(DPM)_2$ from decomposing before

TABLE I Preparation conditions for $NdBa_2Cu_3O_{7-x}$ (NBCO) films and Sr_2AlTaO_6 (SAT) films by MOCVD

	(a) NBCO films	(b) SAT films
Source temperature and carrier Ar flow rate		
	$Nd(TMOD)_3$	$Sr(DPM)_2$
	140–143 °C	200 °C
	50 sccm	30 sccm × 2
	$Ba(DPM)_2$ · Pentaene	$TaAl(O\text{-}iC_3H_7)_8$
	134–137 °C	70–80 °C
	100 sccm ×2	70–140 sccm
	$Cu(DPM)_2$	
	110–115 °C	
	50 sccm	
	dpm-H	
	50 °C	
	50 sccm	
Substrate temperature	820 °C	740 °C
Total gas flow rate	670 sccm	1000 sccm
Deposition pressure	10 Torr	10 Torr
O_2 pressure	1 Torr	1 Torr
Deposition rate	20–25 nm/h	200 nm/h
Thickness	300 nm	300 nm

Present address: Tri Chemical Laboratories Inc., 8154-217 Uenohara, Uenohara-Machi, Kitaturu-Gun, Yamanashi 409-0112, Japan
‡Author to whom all correspondence should be addressed.

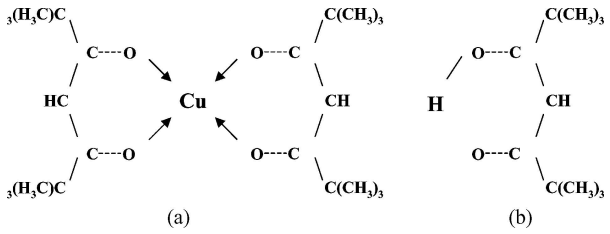

Figure 1 Molecular structures of (a) $Cu(DPM)_2$ and (b) dpm-H.

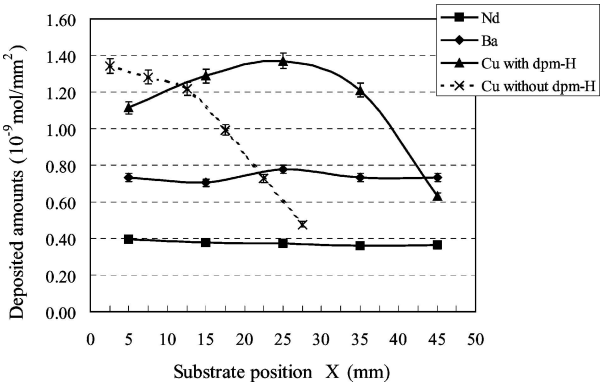

Figure 2 Distribution of deposited amounts of Nd, Ba, and Cu in $NdBa_2Cu_3O_{7-x}$ films on $SrTiO_3$ substrates measured by inductively coupled plasma atomic emission spectrometry.

it reached the substrate. Fig. 1 shows molecular structures of $Cu(DPM)_2$ and dpm-H. The presence of additional organic combine, -dpm, around $Cu(DPM)_2$ probably delays complete decomposition of $Cu(DPM)_2$.

Five STO (100) substrates were set on the substrate holder along the direction of gas flow. Deposition experiments were carried out with and without dpm-H addition. Fig. 2 shows the composition of the deposited NBCO films measured by inductively coupled plasma atomic emission spectrometry. The Cu composition significantly depends on the substrate position, while the Nd and Ba compositions are almost unchanged. Uniformity of Cu distribution is clearly improved by adding dpm-H. Almost homogeneous distribution of all the elements is attained at the substrate position $X = 15$–35 mm. However, the Cu content is slightly higher than stoichiometric. We controlled the flow rate of $Cu(DPM)_2$ and could prepare stoichiometric NBCO films with a thickness of 300 nm. Fig. 3 shows the

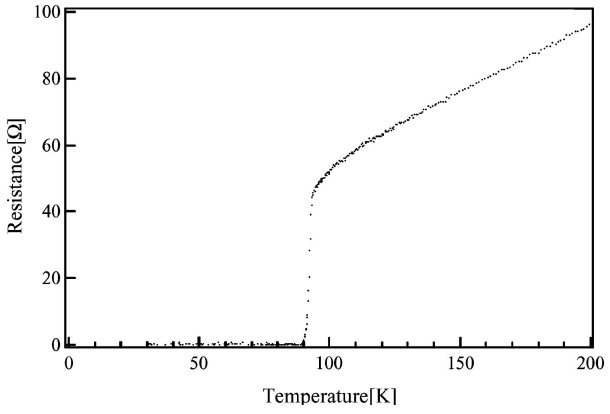

Figure 3 Temperature dependence of the resistance for a nearly stoichiometric $NdBa_2Cu_3O_{7-x}$ film on a $SrTiO_3$ substrate.

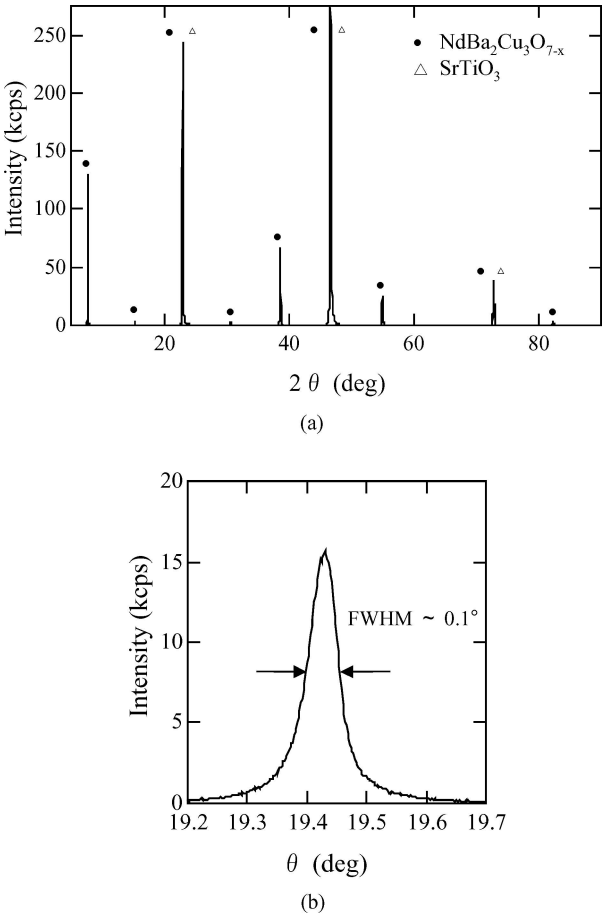

Figure 4 X-ray diffraction patterns for a $NdBa_2Cu_3O_{7-x}$ (NBCO) film on a $SrTiO_3$ substrate: (a) $2\theta - \theta$ scan and (b) Rocking curve of NBCO (005) peak.

temperature dependence of the resistance for an almost stoichiometric NBCO film measured by a four-probe method. The zero-resistance T_c is 90 K. Orientation and crystallinity were examined by X-ray diffraction (XRD). Fig. 4 shows XRD patterns for the NBCO film. The NBCO film shows *c*-axis orientation and the FWHM value of the (005) rocking curve is approximately 0.1°, which is comparable to that of high-quality NBCO thin films prepared by PLD [8, 15]. Fig. 5 shows

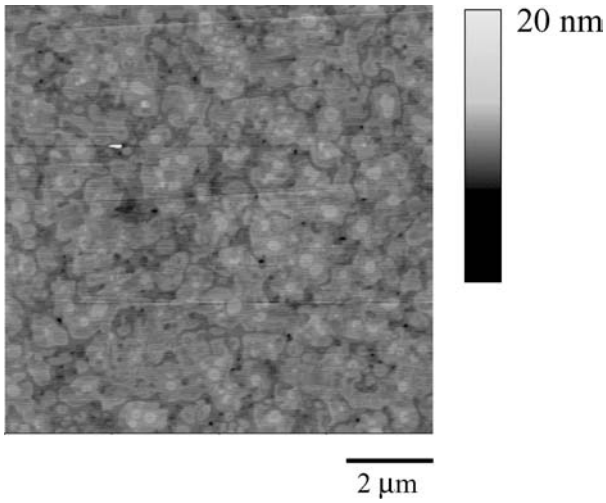

Figure 5 Surface morphology in a 10 μm square area of a $NdBa_2Cu_3O_{7-x}$ film on a $SrTiO_3$ substrate observed by atomic force microscope.

the surface morphology of the NBCO film observed by atomic force microscope. Crystal grains with a size of about 1 μm are tightly connected each other. The root mean square (RMS) surface roughness for the NBCO is 0.7 nm in a 10 μm square area. Thus the obtained NBCO films seem to have sufficiently high quality for applications such as the electrodes of ramp-edge junctions.

Next, we fabricated ramp-edge junctions using the 300-nm-thick NBCO films on STO substrates by an interface-modified process. SAT films were prepared on the NBCO films by the same procedure that was previously reported [13, 14]. Table I (b) lists the preparation conditions for SAT films. The SAT/NBCO bilayer was patterned to fabricate a ramp-edge structure with the angle to the substrate surface 30° by using standard photolithography and Ar ion milling. Then an amorphous layer was formed on the ramp surface by Ar ion irradiation for 90 s at an acceleration voltage of 500 V and the incident angle of 90° to the substrate. A counter NBCO layer was deposited under the same conditions for the base NBCO layer. A 600-nm-thick Au film was sputtered on the counter NBCO layer. Finally, the Au layer and the counter NBCO layer were patterned to a width of 5 μm by photolithography and Ar ion milling. Current-voltage (I–V) characteristics of the junctions were measured by a four-probe method in a magnetic shield.

Among 14 junctions on a chip fabricated with the counter NBCO layer deposited at 820 °C, one junction showed resistively-shunted-junction (RSJ) type I–V characteristics at 4.2 K, as shown in Fig. 6. When a magnetic field was applied, this junction showed critical current (I_c) modulation of about 20%, indicating that interface-modified Josephson junctions can be fabricated by MOCVD. However, flux-flow type I–V characteristics with a typical I_c of several mA were observed in several junctions in the same chip and a majority of junctions exhibited linear I–V characteristics without a supercurrent. Another chip was fabricated with the counter NBCO layer deposited at a slightly lower temperature of 770 °C. The junctions in this chip also showed a large spread of characteristics including RSJ, flux-flow, and resistor types. It should be noted that most of the junctions had similar junction resistance of approximately 1 Ω even for those exhibiting resistor-type curves. The origin of the large spread of characteristics in the present junctions is not clear yet. The existence of resistor-type characteristics suggests that superconductivity at the junction interface was degraded in some regions during the counter-electrode deposition process. Since the deposition time needed for the counter-electrode was longer than 10 hr and the deposition temperature for the counter-electrode was substantially higher than that in other deposition methods such as PLD (typically 670–750 °C), surface contamination or oxygen loss in the base electrode might occur. Further investigation on the fabrication conditions including the ion bombardment conditions, deposition conditions, and the annealing conditions are required to obtain Josephson junctions with lower excess current and a smaller spread in characteristics.

In conclusion, we have attempted fabrication of NBCO ramp-edge Josephson junctions with interface-modified barriers by MOCVD. High-quality c-axis orientated NBCO films with a thickness of 300 nm were successfully deposited. Dpm-H addition to the carrier gas was effective to control the decomposition area of $Cu(DPM)_2$ in the MOCVD reactor and thus we could reproducibly obtain nearly stoichiometric films. The results of the trial fabrication of NBCO ramp-edge junctions indicated that interface-modified Josephson junctions can be fabricated also by MOCVD, though optimization of fabrication conditions are necessary to improve the quality and the homogeneity of junction characteristics.

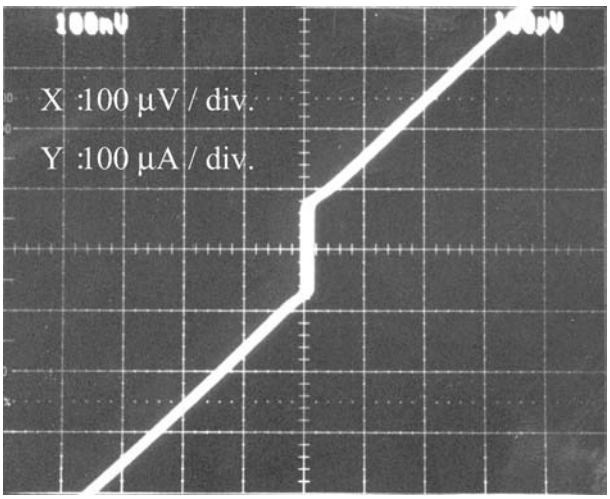

Figure 6 I–V characteristics of a $NdBa_2Cu_3O_{7-x}$ ramp-edge junction with an interface-modified barrier at 4.2 K.

Acknowledgments

The authors would like to thank Y. Takahashi of SHARP Corporation for his advices in preparation of SAT films. This work was supported by the New Energy and Industrial Technology Development Organization (NEDO) as Collaborative Research and Development of Fundamental Technologies for Superconductivity Applications.

References

1. B. H. MOECKLY and K. CHAR, *Appl. Phys. Lett.* **71** (1997) 2526.
2. T. SATOH, M. HIDAKA and S. TAHARA, *IEEE Trans. Appl. Supercond.* **9** (1999) 3141.
3. A. FUJIMAKI, K. KAWAI, N. HAYASHI, M. HORIBE, M. MARUYAMA and H. HAYAKAWA, *ibid.* **9** (1999) 3436.
4. H. KATSUNO, S. INOUE, T. NAGANO and J. YOSHIDA, *Appl. Phys. Lett.* **79** (2001) 4189.
5. Y. ISHIMARU, Y. WU, M. HORIBE, H. WAKANA, S. ADACHI, Y. TARUTANI and K. TANABE, in "Studies of High Temperature Superconductors", edited by A. Narlikar (Nova Sci. Pub. Inc., New York, 2002) Vol. 43, p. 195.
6. H. WAKANA, S. ADACHI, M. HORIBE, Y. ISHIMARU, O. HORIBE, Y. TARUTANI and K. TANABE, *Jpn. J. Appl. Phys.* **41** (2002) L239.
7. J. G. WEN, N. KOSHIZUKA, S. TANAKA, T. SATOH, M. HIDAKA and S. TAHARA, *Appl. Phys. Lett.* **75** (1999) 2470.
8. M. BADAYE, W. TING, K. FUKUSHIMA, N. KOSHIZUKA, T. MORISHITA and S. TANAKA, *ibid.* **67** (1995) 2155.

9. M. KOMATSU, F. WANG, N. TANAKA, H. ZAMA and T. MORISHITA, *J. Cryst. Growth* **205** (1999) 277.
10. Y. KUMAGAI, Y. YOSHIDA, M. IWATA, M. HASEGAWA, Y. SUGAWARA, T. HIRAYAMA, Y. IKUHARA, I. HIRABAYASHI and Y. TAKAI, *Physica C* **304** (1998) 35.
11. C. D. BRANDLE and V. J. FRATELLO, *J. Mater. Res.* **5** (1990) 2160.
12. A. T. FINDIKOGLU, C. DOUGHTY, S. BHATTECHARYA, Q. LI, X. X. XI, T. VENKATESAN, R. E. FAHEY, A. J. STRAUSS and J. M. PHILLIPS, *Appl. Phys. Lett.* **61** (1992) 1718.
13. Y. TAKAHASHI, H. ZAMA, Y. ISHIMARU, N. INOUE, Y. WU, T. MORISHITA and K. TANABE, *Jpn. J. Appl. Phys.* **41** (2002) 590.
14. Y. TAKAHASHI, Y. NAKAJIMA, T. MORISHITA and K. TANABE, *Physica C* **378–381** (2002) 1357.
15. YIJIE LI and K. TANABE, *ibid.* **324** (1999) 198.

Received 27 February
and accepted 29 September 2004