

PREPARATION AND PROPERTIES OF ALL HIGH  $T_c$  SNS-TYPE EDGE DC SQUIDS

J. Gao, W.A.M. Aarnink, G.J. Gerritsma, D. Veldhuis, and H. Rogalla,  
University of Twente, Faculty of Applied Physics,  
P.O. Box 217, 7500 AE Enschede, The Netherlands.

Abstract

High- $T_c$  SNS-type Josephson junctions and dc SQUIDs were successfully fabricated using hetero-epitaxially grown multilayers of  $\text{YBa}_2\text{Cu}_3\text{O}_x$  and  $\text{PrBa}_2\text{Cu}_3\text{O}_x$ . These layers are c-axis oriented and hence edges of the multilayers give rise to a current flow in the ab-plane between the electrodes of a Josephson junction. The necessary structuring was done by Ar ion beam etching. The individual junctions exhibit a supercurrent up to 80 K. The  $I_c R_n$ -product of these junctions usually has a lower limit of 8 mV at 4.2 K. Voltage modulation of the first dc SQUIDs can be observed up to 66 K. At 4.2 K we investigated the voltage modulation for various bias currents and also performed noise measurements. Details on the fabrication and measurements are presented.

Introduction

The discovery of high critical temperature copper-oxide based superconductors has given great impetus to the development of electronic applications of superconductivity. Since their discovery in 1986<sup>1</sup> much effort has been spent on the preparation and study of high  $T_c$  superconducting thin film Josephson junctions. The first high  $T_c$  junction was made at IBM using thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_x$  (YBCO)<sup>2</sup>, rapidly being followed by a number of others<sup>3,4,5</sup>. More recently also junctions in TBCCO<sup>6</sup> and BSCCO<sup>7</sup> materials were fabricated. However, all these junctions depend on the weak-link behaviour of a more or less arbitrary number of naturally occurring grain boundaries in polycrystalline thin films. In order to fabricate junctions with controllable properties several research groups started working on multilayer techniques to create artificial barriers between two superconductors. Both, normal metal as well as insulating barrier materials are used to fabricate SNS or SIS junctions. So far the first result reported in literature of a working SNS-like device was obtained by a group at Bellcore using laser deposited multilayers of YBCO/PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>(PBCO)/YBCO/Au<sup>8</sup>.

It is well known that the YBCO superconductor, having a perovskite structure, is highly anisotropic. The superconducting coherence length perpendicular to the c-axis is much longer than parallel to the c-axis ( $\xi_c(0) \approx 2-3$  Å,  $\xi_{ab}(0) \approx 12-15$  Å). Hence the optimal direction for tunneling is in the ab-plane. Unfortunately nowadays most high quality YBCO thin films are c-axis oriented, although recently also a-axis oriented films of YBCO have been reported<sup>9</sup>. Therefore sandwich type junctions, where current has to be transported in the c-axis direction, would mean tunneling parallel to the c-axis direction, which is supposed to be very difficult. In our laboratory we also tried to make these type of junctions, using off-axis rf magnetron sputter deposition, but for the mentioned barrier layer thickness in ref.8 we could not obtain the Josephson effect. At IBM, YBCO edge junction SQUIDs were fabricated using a barrier made by Ar ion bombardment and a plasma oxyfluoride process. In these type of junctions current transport and tunneling or proximity effect in the ab-plane are utilized. The results are however not very reproducible<sup>10</sup>. Recently we have reported the controlled fabrication of all high  $T_c$  SNS-type edge junctions and dc SQUIDs<sup>11</sup>. In this paper we present the further results on these epitaxial YBCO/PBCO/YBCO Josephson edge junctions and quantum interference devices (SQUIDs). These controllable and reproducible edge junctions show a critical current up to quite high temperatures and could be operated at the boiling temperature of liquid nitrogen.

SQUID Fabrication

In contrast to making a grain boundary junction the materials challenges in making sandwich Josephson junctions between YBCO superconductors are much more formidable. It requires the reliable preparation of high quality epitaxial YBCO thin films. The surface morphology or roughness is of primary concern. Of the electrical properties both high critical temperature and high critical current density are essential. In addition, deposition of a single superconducting layer is not sufficient. It is necessary to prepare epi-heterostructures that involve the successive deposition of several thin epitaxial layers which must be superconducting, insulating or metallic. The difficulty is to make a barrier that (i) survives the high temperature deposition process for the top electrode, (ii) is sufficiently thin, i.e. a few nm, but without pinholes to have a usefully high Josephson tunnel current density and (iii) can allow the epitaxial growth of the top YBCO layer. Our high  $T_c$  Josephson junctions have been fabricated from hetero-epitaxially grown multilayers. The multilayer structures include both thin barriers for junctions and insulating layers between the YBCO superconductors. A PBCO layer was used as normal conducting barrier layer in the SNS edge sandwich because of its isomorphic structure and because its lattice constant is similar to YBCO<sup>12</sup>. Details about the preparation of YBCO thin films and YBCO / PBCO / YBCO multilayers have been published in previous papers<sup>13,14</sup>. Briefly, the YBCO and PBCO thin films have been deposited in situ by a modified off-axis rf magnetron sputter technique. A ground plate was mounted, facing the target at a 45° angle. This geometry greatly reduces backsputter effects and similar to ion beam sputtering, the sputter parameters are much less interdependent. Stoichiometric sintered YBCO and PBCO targets with a diameter of 50 mm were used. As substrates, we used (100) surfaces of Yttrium-stabilized ZrO<sub>2</sub>. The sputter gas was a mixture of argon and oxygen which consisted, in most cases, of 50% oxygen and 50% argon. Typical sputter gas pressures are between  $5 \times 10^{-2}$  and  $3 \times 10^{-1}$  mbar. During deposition the substrates were heated to temperatures between 640 and 700 °C. The incident rf-sputter power density was 2-5 W/cm<sup>2</sup>.

By means of this technique, we routinely obtain transition temperatures,  $T_{c,zero}$ , of about 90 K and critical current densities at 77K are found to be higher than  $10^6$  A/cm<sup>2</sup>. For the YBCO / PBCO / YBCO multilayers the superconducting properties are not affected by the presence of the PBCO layer. On X-ray diffraction patterns, besides the substrate reflections, only (00l) peaks are present. The full width at half maximum of the (005) peak was observed to be 0.35°. It is somewhat broader than that we found for single films (for YBCO thin films:  $\Delta\theta_{50} = 0.28^\circ$ , for PBCO thin films:  $\Delta\theta_{50} = 0.30^\circ$ ), but still indicates that these multilayers are highly textured. This is confirmed by He-ion channeling on a trilayer and Auger sputter profiling. Within the depth resolution limits of RBS of about 5 nm, no interdiffusion between layers could be observed.

In order to fabricate Y/P/Y junctions in the ab-plane we used a two step process. In the first step, a YBCO layer with a thickness of 30-200 nm and a PBCO toplayer have been grown in situ. This PBCO layer is relatively thick (around 150-250 nm) in order to avoid Josephson coupling between two YBCO layers along the c-axis direction. This however gives rise to resistive shunting between the two superconducting electrodes. So for some samples an extra ZrO<sub>2</sub> layer was used as insulator. The ZrO<sub>2</sub> insulating layer was deposited by ion beam sputtering. A focused ion gun of 3 cm diameter was used as ion source and argon at a pressure of  $6 \times 10^{-4}$  mbar was used as sputter gas. As target we used yttrium stabilized ZrO<sub>2</sub> ceramic material. The voltage and current of the Ar<sup>7</sup> ion beam are 800

Manuscript received September 24, 1990.

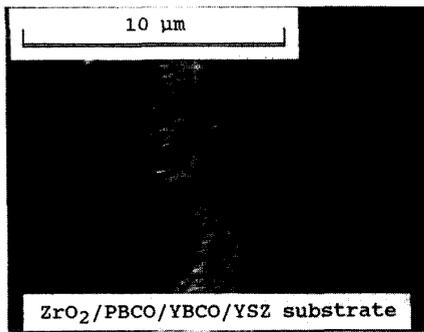


Figure 1. SEM micrograph of a base electrode edge before deposition of the counter electrode.

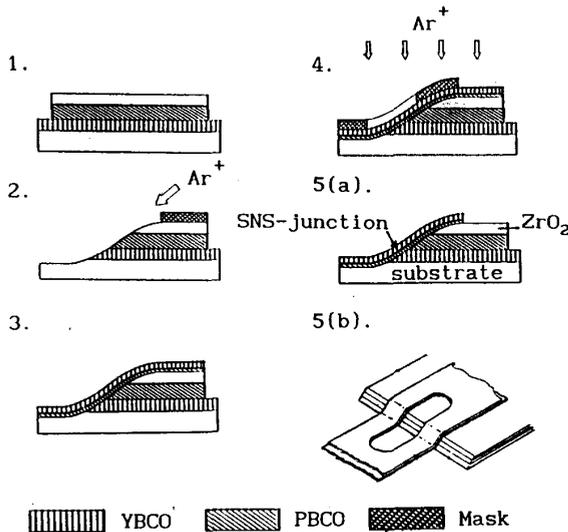


Figure 2. Fabrication sequence for the edge SQUIDS: 1) In situ growth of the  $\text{YBa}_2\text{Cu}_3\text{O}_x$  bottom layer,  $\text{PrBa}_2\text{Cu}_3\text{O}_x$  and insulating  $\text{ZrO}_2$ . 2) Fabrication of the base electrode edge by the Ar ion beam etching. 3) Deposition of the  $\text{PrBa}_2\text{Cu}_3\text{O}_x$  barrier and  $\text{YBa}_2\text{Cu}_3\text{O}_x$  top electrode. 4) The sample is lithographically patterned and ion milled through to make the loop. Finally four silver electrodes are deposited by sputtering. 5) Schematic of a finished edge dc SQUID. (a) the cross-section view and (b) the perspective review.

V and 20 mA, respectively. The resistance of this insulating layer is more than  $10 \text{ M}\Omega$  for a  $16 \text{ mm}^2$  surface area. In this case, even though a higher decrease of transition temperature occurs, perhaps due to interdiffusion of  $\text{ZrO}_2$ , a zero resistance temperature above 80 K is still being achieved. Then a part of the multilayer is covered by a photoresist stencil and the unprotected area was etched by an Argon ion beam with an incident angle of 45-65 degree in order to fabricate an edge. Our ion beam parameters are typically 3.5 kV acceleration voltage and  $30 \mu\text{A}/\text{cm}^2$  beam current density at the sample. Figure 1 is the SEM micrograph of a base electrode edge before deposition of the counter electrode.

After removing the photoresist stencil a thin PBCO barrier layer and YBCO top layer are deposited. As the edge may be contaminated in between the two processes we cleaned the edge with the Ar ion beam before depositing the barrier layer. Next a junction or a dc SQUID is formed across the edge using photo lithographic patterning and ion beam etching.

The resulting edge junction cross section is shown in figure 2-(5). Finally the lateral dimension of the edge is reduced to obtain junctions with a width ranging between 10 and 1000  $\mu\text{m}$ . The height of these junctions are typically between 30 and 100 nm. In all we fabricated about 20 junctions, each on a different substrate and about 6 dc SQUIDS. The typical transition temperature is around 80 K for the junctions.

#### Measurements

In figure 3 we present the I-V-characteristics of one of these shunted junctions, i.e. without insulating  $\text{ZrO}_2$  layer. The PBCO barrier layer thickness was approximately 6 nm and the supercurrent vanishes around 80 K. This is about 10 K below the transition temperature of our YBCO thin films or YBCO/PBCO/YBCO multilayers. The critical current density of our best junction is approximately  $1000 \text{ A}/\text{cm}^2$  at 77 K. The  $I_c R_n$ -product of this junction at 4.2 K is around 8 mV.

In figure 4 we plotted the critical current  $I_c$  vs.  $1-T/T_c$  on a double logarithmic scale. From this we obtain that the critical current is proportional to  $(1-T/T_c)^2$ . This is characteristic of a SNS-junction<sup>15</sup>. The evidence is not conclusive however. Due to the depression of the order parameter at an interface the same temperature dependence may be found for SIS junctions of high  $T_c$  superconducting

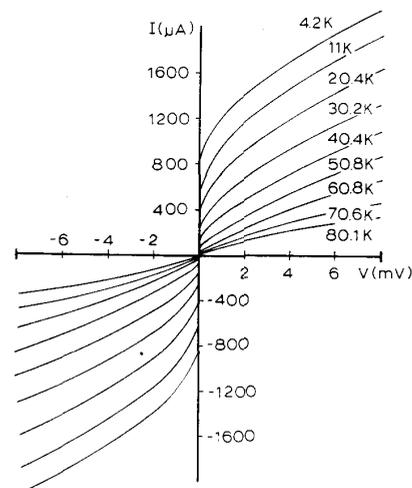


Figure 3. Current-voltage characteristics of one of our edge junctions without  $\text{ZrO}_2$  layer at different temperatures. This junction is shunted.

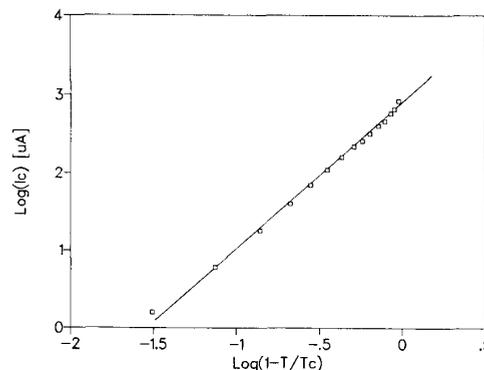


Figure 4. The critical current  $I_c$  as function of  $(1-T/T_c)$  for the junction of fig.2. A slope of two has been observed. This indicates SNS behaviour.

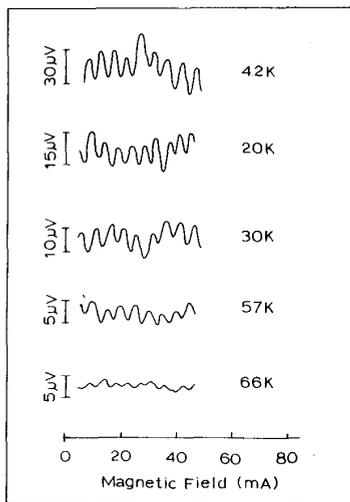


Figure 5. Voltage vs magnetic field for one of the edge junction dc SQUIDs at different temperatures. The magnetic field is plotted in units of current through the magnetic coil and 20 mA current yields approximately 7 flux quanta in the loop.

materials<sup>16</sup>. As we have, however, also fabricated junctions with barrier layer thickness up to 30 nm, being much too thick for tunneling, we conclude the junctions are indeed of the SNS type.

For the fabrication of the dc SQUIDs a hole of  $5 \times 150 \mu\text{m}^2$  was etched across the edge using photo lithography and ion beam etching. In order to minimize the probability of having shorts between the junction electrodes, we used a barrier layer thickness of 20 nm. Furthermore only  $\text{ZrO}_2$  was used to separate the top and bottom electrode in order to prevent resistive shunting of the junction and hence to maximize the dc SQUID voltage modulation.

In figure 5 we present the flux voltage characteristics of one of our dc SQUIDs in the temperature range from 4.2 K to 66 K. The height of this edge junction is 120 nm whereas the width is  $15 \mu\text{m}$  and the critical current density at 4.2 K is approximately  $150 \text{ A/cm}^2$ . The normal state resistance is about

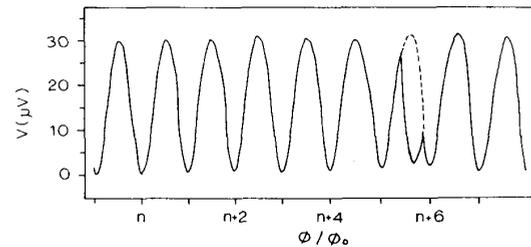
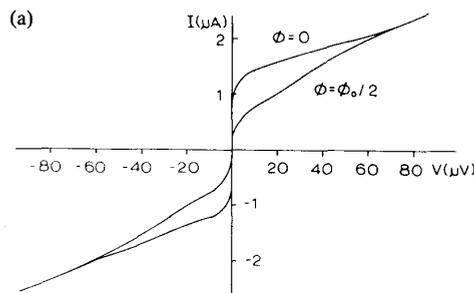


Figure 7. An example of the output voltage modulation curves which shows a phase jump among the normal periodic peaks. Structures like this always appeared with exactly the same applied flux.

$20 \Omega$  and is almost independent of temperature. The  $V-\Phi$  curves are well periodic up to an applied flux of several tens of flux quanta. Even above 66 K some modulation could be observed, but the signal is too small and noisy for practical applications. It should however be noted that these measurements were carried out in an unshielded environment. The modulation period corresponds to approximately one flux quantum in the SQUID loop and does not change between 4.2 K to 66 K. The total modulation depth  $m = \Delta V_m / I_c$  is roughly 20% of the critical current and the modulation voltage at 4.2 K and 66 K are typically  $25 \mu\text{V}$  and  $2 \mu\text{V}$ , respectively. The  $LI_c$ -product is roughly  $0.2\Phi_0$  as calculated from the geometrical data of the SQUID.

Figure 6 shows a set of typical  $V_{\text{mod}}$  vs.  $\Phi$  characteristics of a SQUID for various bias currents measured at 4.2 K in a well shielded environment. The curves are very smooth in the usual bias regime  $I_B \approx I_c \cdot 2.5I_c$ . The maximum voltage modulation has been obtained for  $I_B \approx 1.3 \mu\text{A}$  to be approximately  $25 \mu\text{V}$ . Compared to the other results obtained on single-level or multi-level high  $T_c$  SQUIDs reported so far<sup>3,4,5,8,9</sup> this maximum voltage modulation is quite large: in normalized units  $\Delta V_m / I_c \cdot R_n \approx 0.35$ . No significant changes in the voltage modulation data were seen even after 6-8 cryogenic/room temperature cycles, showing the relatively robust nature of these edge junctions.

The well periodic modulation peaks can be observed until quite high applied field. However phase jumps sometimes are present. Figure 7 is an example of the output voltage modulation curves which shows a phase jump among the normal periodic peaks. Structures like this always appeared with exactly the same applied field. They are attributed to the

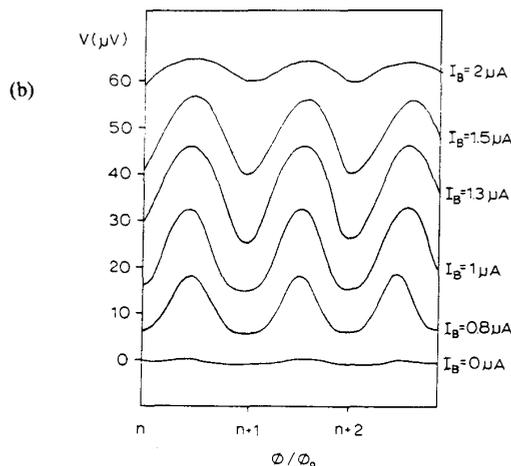


Figure 6. (a) Characteristic of a dc SQUID measured at 4.2 K. (b) A set of typical  $V_{\text{mod}}$  vs.  $\Phi$  characteristics of this SQUID for various bias currents measured at 4.2 K.

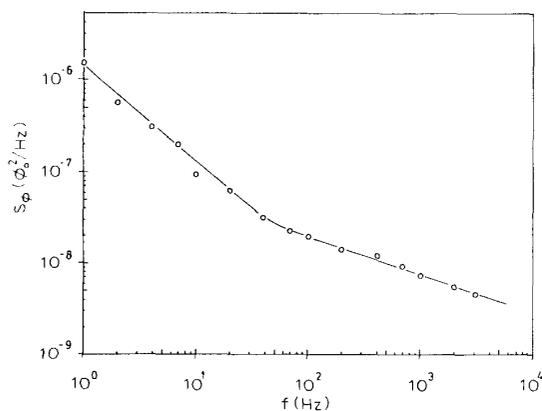


Figure 8. The noise spectra of one of our SQUIDs measured at 4.2K.

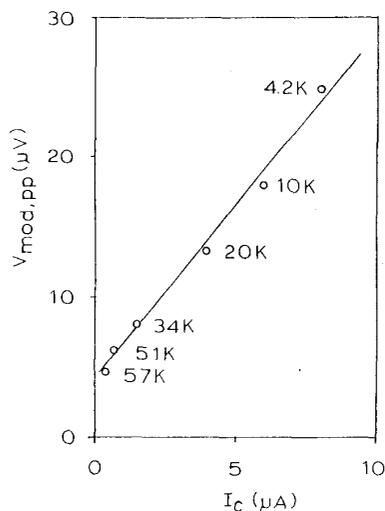


Figure 9. The modulation voltage as function of the critical current.

sudden movement of vortices across the SQUID loop at certain applied fields where a vortex jumps from one pinning site to another.

The noise power spectrum of this particular SQUID was measured at 4.2 K and is given in figure 8. It shows an increased noise level at frequencies below about 40 Hz (at 40 Hz the flux noise was observed to be  $1.8 \times 10^{-4} \Phi_0/\text{Hz}^{1/2}$ ) having a  $1/f$  frequency dependence of  $S_\phi(f) \approx 2 \times 10^{-8}/f \Phi_0^2/\text{Hz}$ . This noise is caused probably by the jumping of vortices. In real time on our oscilloscope screen, we observed jumping between two different base line voltage levels as is typical for telegraph noise. The remaining variations around these two base lines are one order of magnitude lower than the base line jumps. Compared to the noise of classical dc SQUIDs the noise of high  $T_c$  SQUIDs is higher, but it is low enough to make these high  $T_c$  oxide superconductor SQUIDs quite useful in many applications. Furthermore we think that by a better design of the dc SQUIDs vortex jumps may be avoided and hence the resulting telegraph noise reduced.

The temperature dependence of the modulation voltage has been studied as well as the critical current. A linear dependence of modulation voltage on critical current has been observed (figure 9). Although up to now the practical operation temperature of these first SQUIDs is still lower than the boiling temperature of liquid nitrogen there seems to be no difficulty to obtain useful SQUIDs which could be operated at 77 K in the near future. The important point is that the supercurrent of these artificial barrier edge junctions flows through a well controlled PBCO layer in the ab-plane of the YBCO layer. It gains a remarkable advantage over the usual multilayer junctions in terms of reproducibility and controllability. Furthermore it should be noted that we could increase the bias current of our dc SQUID up to 1 mA at 4.2 K without any damage to the junctions which could be seen in the I-V characteristics. From this we conclude that the Josephson effect is not caused by shorts across the barrier layer as was found for the edge junctions of Laibowitz et al<sup>10</sup>.

### Conclusions

In summary, we have successfully fabricated artificial barrier edge junctions and dc-SQUIDs of YBCO material with a good reproducibility. These junctions are based on high  $T_c$  epi-heterostructures in which the superconducting layers still keep good superconducting properties. By varying the PBCO barrier layer thickness the current through the edge junctions can be controlled independently from the junction height and width. Voltage modulation with applied flux has been observed up to 66 K. We think such edge junction SQUIDs will provide genuine applications for high  $T_c$  superconductors, especially where low critical current and small capacitances are required.

We acknowledge the help of B.B.G.Klopman, D. J. Adelerhof, and A.J.H.M. Rijnders. This work was supported by the Dutch National Research Program on High  $T_c$  Superconductors.

### References

1. J.G.Bednorz and K.A.Müller, *Z. Physik*, **B64**, 189, 1986.
2. R.H.Koch, C.P.Umbach, G.J.Clark, P.Chaudhari, and R.B.Laibowitz, *Appl. Phys. Lett.*, **51**, 200, 1987.
3. J.E.Zimmerman, J.A.Beall, M.W.Cromar, and R.H.Ono, *Appl. Phys. Lett.*, **51**, 617, 1987.
4. B.Häuser, M.Diegel, and H.Rogalla, *Appl. Phys. Lett.*, **52**, 844, 1988.
5. I.Takeuchi, J.S.Tsai, H.Tsuge, and N.Matsukara, *Jan. J. Appl. Phys.*, **27**, 2265, 1988.
6. R.H. Koch, W.J. Gallagher, B. Bumble, and W.Y. Lee, *Appl. Phys. Lett.*, **54**, 951, 1989.
7. D.W.Face, J.T.Kacera, D.G.Steel, J.M.Graybeal, T.P.Orlando, and D.A.Rudman, Preprint.
8. R.H.Koch, W.J.Gallagher, V.Foglietti, B.Oh, R.B.Laibowitz, G.Koren, A.Gupta, and W.Y.Lee, Preprint.
9. C.B. Eom, A.F. Marshall, S.S. Laderman, R.D. Jacowitz and T.H. Geballe, to published in *Science*.
10. R.B.Laibowitz, R.H.Koch, A.Gupta, G.Koren, W.J.Gallagher, V.Foglietti, B.Oh, and J.M.Viggiano, *Appl. Phys. Lett.*, **56**, 686, 1990.
11. J. Gao, W.A.W. Aarnink, G.J. Gerritsma, and H. Rogalla, "Controlled Preparation of All High  $T_c$  SNS-type Edge Junctions and Dc SQUIDs", to be published in *Physica C*.
12. U.Poppe, P.Prieto, J.Schubert, H.Solner, K.Urban, and C.Buchal, *Solid State Comm.*, **71**, 569, 1989.
13. J. Gao, B. Häuser, H. Rogalla, "High Critical Current Density Thin and Ultra Thin Films of  $\text{YBa}_2\text{Cu}_3\text{O}_x$  Made by a Modified Rf-magnetron Sputter Technique", *J. Appl. Phys.*, **67**, 2512, 1990.
14. J. Gao, W.A.W. Aarnink, G.J. Gerritsma, H. Rogalla, "High Quality  $\text{YBa}_2\text{Cu}_3\text{O}_x$  Ultra Thin Films and YBCO/PBCO/YBCO Multilayer Films by a Modified Off-axis Sputtering", to be published in *Appl. Surface Science*.
15. J.Clark, *Proc. Roy. Soc. A*, **208**, 447, 1969.
16. D. Deutscher and K.A. Müller, *Phys. Rev. Lett.*, **59**, 1745, 1987.