Ramp type HTS Josephson junctions with PrBaCuGaO barriers

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Abstract—**Ramp type Josephson junctions have been fabricated using $DyBa_2Cu_3O_{7-\delta}$ as electrode material and $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ with x=0, 0.10 and 0.40 as junction barriers. Barrier thickness lie between 6-30 nm. Several junctions without barrier were made in order to find ways to minimize the damage of the ramp interface. In total about 40 chips were fabricated each containing several junctions and their I-V characteristics measured for various temperatures down to 4.2 K. Only those junctions showing clear RSJ-like curves were selected to be analyzed. In some cases we also measured I_c as a function of a small applied field and obtained a clear Fraunhofer pattern, but there is a tendency to flux trapping as evidenced by LTSEM. It was found at 4.2 K that the critical current density J_c scales with the specific resistance R_nA as $J_c=C_{bar}(R_nA)^{-m}$ (m=1.8±0.5). The barrier material dependent constant C_{bar} increases with x, whereas, for a given d, J_c is constant and R_nA increases.**

I. Introduction

At present practical HTS Josephson junctions have reduced I_cR_n values of the order 0.1-1 mV compared to the gap voltage (~20 mV). Reasons for the reduction in I_cR_n may be interface effects [1],[2], normal state-shunting [3], an inelastic transport process or coulomb blockade of resonant tunnel channels [4].

The ramp type junction structure [5]-[9] allows adjustment of junction properties by changing the barrier thickness or the barrier material itself. The barrier material has to be grown epitaxially on an etched ramp in a (001) oriented $DyBa_2Cu_3O_{7-\delta}$ film to allow deposition of a high quality top electrode. Still then the barrier material should be carefully chosen with respect to materials properties [10], [11]. A large difference in thermal expansivity of the barrier material will induce strain that will relax at the interface by formation of defects in the electrodes, thus causing an interface resistance dominating the barrier properties if combined with a low bulk resistivity. The material may enhance formation of impurity phases like Dy_2O_3 resulting in a strongly modulated current pattern (and with it a large spread in junction parameters). It is preferred to use a 123 related material like $PrBa_2Cu_3O_{7-\delta}$, for its high bulk resistivity and structural similarity with $DyBa_2Cu_3O_{7-\delta}$, showing small interdiffusion and interreaction and the similar way of chain loading with oxygen during cooldown. $PrBa_2Cu_3O_{7-\delta}$ is the only 123 material that does not show a transition to superconductivity and is not even metallic. The reason for this dissimilar behavior is still matter of discussion [12], [13]. In this material, although there is chain oxygen loading, this does not lead to depletion of carriers from the plane layers [14] nor to a destruction of the anti-ferromagnetic ordering. The ordering into long Cu-O chain segments causes a tetragonal to orthorhombic transition around $7-\delta=6.35$ [16] like it does for the other 123 materials. The chain layer conductivity appears to be dominant in $PrBa_2Cu_3O_{7-\delta}$, considering the dramatic decrease in resistivity by rising the oxygen content [15].

As indicated by numerous neutron-diffraction studies of transition-metal doped $RBa_2Cu_{3-x}M_xO_{7-\delta}$, where R={Y or a rare earth}, M={Fe, Al, Co, Ga} preferentially substitute the chain Cu(1) atoms and M={Zn, Ni} the plane Cu(2) atoms (Fe also shows some Cu(2) substitution at higher doping levels) [17], [18]. Probably some mixed substitution for all elements cannot be excluded. In all cases the T_c is systematically suppressed, and the resistivity is strongly enhanced. The long-chain ordering disappears because of oxygen redistribution around the impurity atoms, and results in a crossover to the tetragonal phase between x=0.10-0.20 [19].

Substitution of copper in $PrBa_2Cu_3O_{7-\delta}$ by {Ga, Co and Nb} has been found to result in a tremendous increase in bulk resistivities [20]-[22]. In this paper we report our study of ramp type junctions with $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ (x=0, 0.10 and 0.40) as barrier materials, with barrier thickness varying from 6-30nm, aiming to find out systematics in the junction parameters.

II. Experimental

Target materials of $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ (x=0.10 and 0.40) have been prepared using citrate synthesis [23]. In short: optimal mixture of the elements before reaction is achieved by bringing the starting materials (Pr_6O_{11}, $BaCO_3$, CuO, Ga) in acidic solution and mixing them in the desired stoichiometry along with citric acid. After neutralization with ammonia the solution is evaporated. By performing pyrolysis a polymerization reaction gives, due to the large gas exhaust, a sponge-like structure consisting of particles with a typical size of 50-100 nm. A calcination reaction in flowing oxygen at 800°C burns out all carbon. Then the solid state reaction is carried out at 930°C during 15 hours in flowing oxygen. After grinding and pressing into 5 cm diameter pellets, targets are sintered at 930°C for another 10 hours. The target densities

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obtained are about 80% of the theoretical value. X-ray fluorescence analysis shows the expected material stoichiometries within the accuracy of the measurement (~1%). Formation of the right 123 phase (with the expected tetragonal phase for x=0.40 [20]) is revealed by X-ray diffraction (XRD), and only very minor reflections of barium cuprate just above the noise level can be seen.

Epitaxial films of $YBa_2Cu_3O_{7-\delta}$, $DyBa_2Cu_3O_{7-\delta}$ and $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ (x=0, 0.10 and 0.40) have been deposited with [001] perpendicular to the surface of (001) $SrTiO_3$ substrates using off-axis RF magnetron sputtering from single stoichiometric targets. Substrates are glued with Ag-paint to a stainless heater block and heated to deposition temperature (770°C) until a background pressure of $1x10^{-4}$ Pa is reached. The high deposition temperature is chosen to completely suppress the formation of a-axis oriented material as has been revealed by XRD-measurements. Plasma is created in a 10-20 Pa mixture of 50% Ar and 50% O_2, with traces of H_2O gas (~1‰) for higher reactivity. After 20 minutes presputtering the deposition is started by moving the heater block into position. Deposition rates are typically 70-90 nm/hr, depending strongly on the pressure and heater position. After deposition the samples are cooled down within 60 minutes to room temperature in a 200 mbar O_2 atmosphere.

To rule out Ga-diffusion in the ab-plane we sputtered in-situ a bilayer (using our standard settings for c-axis oriented growth) on a (103) oriented $SrTiO_3$ substrate of 200 nm $PrBa_2Cu_{2.60}Ga_{0.40}O_{7-\delta}$ / 40 nm $DyBa_2Cu_3O_{7-\delta}$. The layers grew coherently tilted (with the [001] rotated over 18° with respect to the substrate normal), as was indicated by X-ray diffraction using a Philips MRD diffractometer. In this way the CuO_2 planes of the bilayer came in direct contact. The T_c of the top layer was 90.5 K, indicating no diffusion of Ga in the crystalline phase. The lattice parameters of the $PrBa_2Cu_{2.60}Ga_{0.40}O_{7-\delta}$ layer corresponded to the right doping level. This agrees well with the findings of Contour et al. for c-oriented multilayers [24], that Ga shows little tendency to diffuse once incorporated in the crystal lattice, even at high temperatures (770°C).

Fabrication of junction structures is performed using a standard photolithography process. After in-situ deposition of the first bilayer of 100 nm $DyBa_2Cu_3O_{7-\delta}$ base electrode and 100 nm $PrBa_2Cu_3O_{7-\delta}$ insulation layer, ramps are structured by Kaufmann Ar-ion beam etching under an angle of 35° with the substrate surface, resulting in ramp angles of 18-22°. The resist is stripped by ultrasonic stirring in acetone and rinsing in 2-propanole. Before deposition of the second bilayer the sample is cleaned by removing a surface layer of 20 nm to minimize the contamination effects induced by contact with the ambient and photoresist in the ex-situ process. After outgassing in the sputter atmosphere, a thin barrier layer (6-30 nm) of $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ and 100 nm top electrode of $DyBa_2Cu_3O_{7-\delta}$ are deposited by the usual procedure.

The length of the overlap, where the top electrode extends over the insulation layer is minimized to 3-5 microns, again by ion beam milling. Metallization is performed by a lift-off process by DC-sputtering of 8 nm Ti adhesion layer and 100 nm Au contact layer. The junctions (5-25 microns wide) are defined in the final etching step. Electrical contacts for measurements are made by mounting the sample on a PC-board and ultrasonic bonding of 25μm Al-wire on the gold pads.

Standard I-V measurements are performed down to 4.2 K in a temperature controlled flow-cryostat with μ-metal shielding. Magnetic field dependence is studied in an unshielded environment with the field applied by an external Helmholtz coil set.

III. Results

Bulk polycrystalline $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ shows a dramatic increase of the resistivity with Ga doping, up to 8 orders of magnitude at 77 K for x=0.40 with respect to undoped $PrBa_2Cu_3O_{7-\delta}$, which is consistent with the values found by Xu et al. [20]. The film resistivities are systematically lower and are increasing at a lower rate with decreasing temperature than the bulk samples [24], [25]. The resistivity at 77 K of thin $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ films increases by 3 orders of magnitude for a Ga doping level x=0.40. Fits to the Mott's variable range hopping model [26] are rather poor, especially for thin films.

In Fig. 1 the critical current density of an Ar-ion beam cleaned ramp contact (without extra $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barrier layer) is compared to that of a bridge in a single layer of $DyBa_2Cu_3O_{7-\delta}$. This is a measure for the quality and the amount of damage that has been induced by performing the ex-situ process of ramp structuring. There is a reduction in J_c of a factor of four to five by the cleaning procedure.

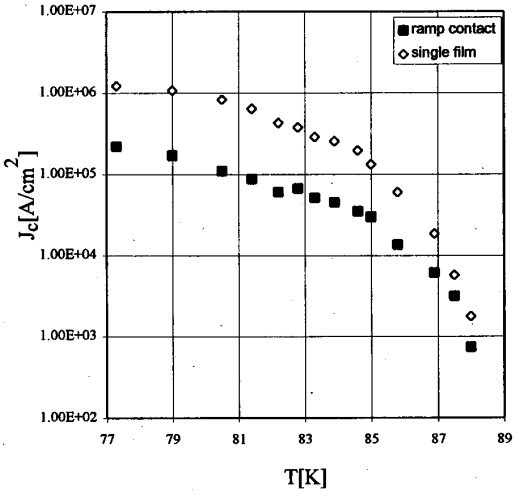

Fig. 1. Critical current densities vs. temperature of a ramp contact (without extra $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barrier layer) compared to that of a bridge in a single layer of $DyBa_2Cu_3O_{7-\delta}$.

We fabricated about 40 chips each containing several junctions. Only those junctions showing clear RSJ-like I-V curves, with low excess current, and straight resistive branches for low bias-conditions were selected. The on-chip spread could usually be explained by the systematic spread in deposition rate across the $1cm^2$ substrate. There is some capacitive loading depending on the length of the overlap area, that leads to hysterisis in the I-V curves. The normal resistance R_n (taken at a few mV) is increasing upon lowering the temperature. This temperature dependence is stronger for larger barrier thickness and is intimately related to the bulk barrier material in the junctions. The barrier thickness is estimated from the sputter time, by using the deposition rate in the [001]-direction, which is frequently determined from step heights in etched films.

About 20% of the structures show strong deviations from this behavior, usually having very high I_c values even for temperatures close to the T_c of the electrodes, very rounded curves indicating flux flow, and no magnetic field modulation of the critical current. This occurs most frequently for thinnest barriers (6-8 nm), presumably because of leaks through pinholes in the barrier layer. For 30 nm thick barriers no superconductive coupling is observed anymore.

In Fig. 2, I-V characteristics of a junction with a 10 nm thick $PrBa_2Cu_3O_{7-\delta}$ barrier at several temperatures is shown. The barrier resistivity is decreasing for increasing temperature. The inset shows the $I_c(B)$ dependence of a junction, measured in an unshielded environment using an external Helmholtz coil with the magnetic field parallel to the barrier plane, piercing the junction from aside. The sin(x)/x fit to the measurements is shown with a solid line. The measurements were done at 56 K, the T_c of both electrodes was 89 K and the estimated barrier thickness in the [001] $PrBa_2Cu_3O_{7-\delta}$ direction is 10±2 nm. Complete suppression of the supercurrent is not always observed in our $I_c(B)$ measurements, typical modulation depths are only 75%.

The effect of Ga doping on the junction parameters can be seen clearly from Fig. 3; in this diagram, the two lines with slope -1 depict constant voltages of 1 mV and 10 mV respectively. The J_c to R_nA characteristics of real tunnel junctions would be parallel to these lines, following the gap voltage. Clearly the solid lines, following J_c vs. R_nA of the junctions with $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barriers, disagree with this behavior. Instead the lines are better fitted by: $J_c=C_{bar}(R_nA)^{-m}$ (m=1.8±0.5), while C_{bar} increases with x, as the lines are shifting upwards for higher doping levels. In Fig. 4, J_c is plotted vs. the estimated barrier thickness d. Within the accuracy of this estimation (d±12%), J_c appears to be largely independent of the Ga-doping level x.

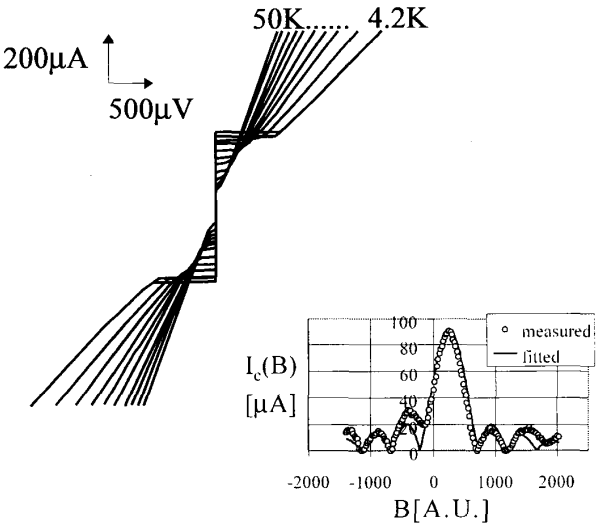

Fig. 2. I-V characteristics of a junction with a 10 nm $PrBa_2Cu_3O_{7-\delta}$ barrier at several temperatures. Inset: $I_c(B)$ dependence of a junction and a sin(x)/x-fit.

IV. Discussion and Conclusions

Ga doped $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ was formed having the right stoichiometric composition and crystal structure. The Ga that is incorporated in the lattice is found to be quite stable, showing no significant out-diffusion even at elevated temperatures (770°C). This is not very surprising as the ions are kept in their lattice position by a strong Coulomb interaction [24].

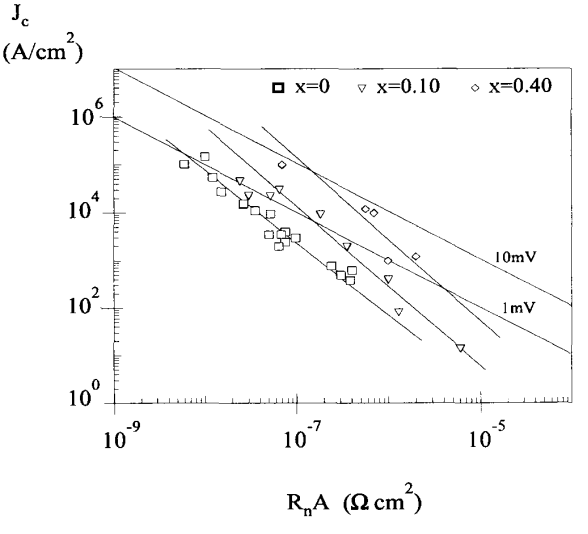

Fig. 3. J_c vs. R_nA for junctions with $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barriers at 4.2 K.

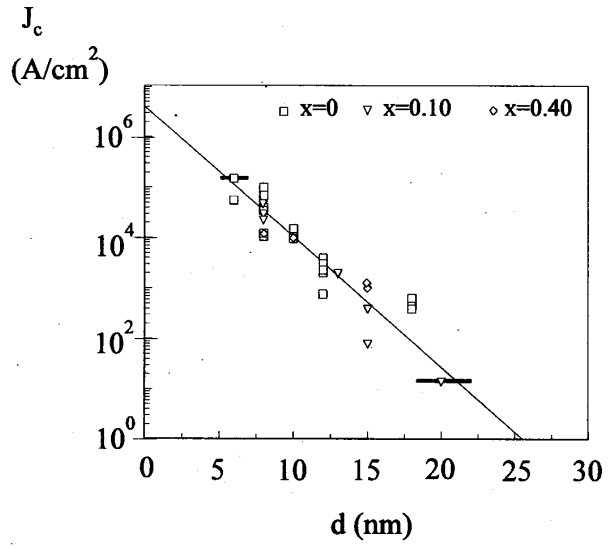

Fig. 4. J_c vs. d for junctions with $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barriers at 4.2 K.

For a number of junctions we measured the $I_c(B)$ dependence in an unshielded environment. They generally have a periodic modulation depth of about 75%. For a few devices complete suppression is observed, with clear deviations from the ideal Fraunhofer pattern though (inset of Fig. 2), presumably due to flux trapping. It was shown using LTSEM measurements, that the ramp type structure might enhance the probability for flux trapping at the ramp [27].

The conduction mechanism in $PrBa_2Cu_3O_{7-\delta}$ is not clear but a kind of hopping transport via localized states is likely [20]. If a barrier layer between two superconducting banks is thin enough it carries supercurrent until the critical current density J_c is reached. In this work it is observed that J_c is largely independent of doping level x; this means that the gain in the I_cR_n value is caused by the increase in the R_nA product. The I_cR_n values are reduced with respect to the gap voltage, and this deviation increases for higher barrier thickness. A resonant tunneling mechanism via localized states seems to be a reasonable option [4] because the transport of Cooper pairs and quasiparticles through a $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barrier have different dependencies on charge trapped in localized states.

From Fig. 4, the fit $J_c \sim e^{-d/\xi}$ gives $\xi \approx 2$nm for the characteristic decay length of the supercurrent with the thickness of $PrBa_2Cu_{3-x}Ga_xO_{7-\delta}$ barriers if we take the thickness d in the (001) direction or some 4-5nm if the thickness is taken parallel to the CuO_2 planes. This is still a much shorter value than the values found by most other authors ([15] and references therein).

The consensus is that the chain and the plane layers have their own role in the conduction in 123 materials. We doped the chain layers of $PrBa_2Cu_3O_{7-\delta}$ with Ga, and the effect on the superconductive coupling was negligible when this material was used for barrier layers in ramp type junctions. The resistivity increased up to an order of magnitude for the same barrier thickness, however. This motivates further research on barrier layers with $PrBa_2Cu_{3-x}M_xO_{7-\delta}$, where M substitutes the plane Cu atoms.

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