

# Grain boundary engineering with gold nanoparticles

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**Abstract.** We investigated high- $T_C$  grain boundary Josephson junctions with and without incorporated gold nanoparticles. Pulsed laser deposition was used for the deposition of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films on  $\text{SrTiO}_3$  bicrystal substrates with different grain boundary angles. During the deposition process, single-crystalline nanoparticles self-assembled from a thin gold layer which was sputtered on the substrate before the YBCO deposition. The interaction between nanoparticles and thin film growth significantly influences the quality of the YBCO films [1]. The critical current density and the critical temperature of the superconducting films can be increased in a defined manner. Furthermore, the nanoparticles influence the growth conditions in the region of the grain boundary and thus the properties of the later patterned Josephson junctions. The comparison between Josephson junctions with and without nanoparticles on the same substrate shows a reduction of the critical current  $I_C$  and an increase of the normal state resistance  $R_N$  for all investigated types of grain boundaries in the areas with gold nanoparticles. In some cases we even found an increase of the resulting  $I_C R_N$  product. We present the influence of light irradiation on the properties of the Josephson junctions.

## 1. Introduction

Josephson junctions are the basic elements of superconducting analogue and digital devices, e.g. SQUIDs and voltage standards. For practical applications it is necessary to develop techniques which allow the preparation of Josephson junctions with specific superconducting properties. Critical current density  $J_C$  and specific normal resistance  $\rho_N$  as well as the capacitance  $C$  of Josephson junction should be optimized independent from each other. In this paper we describe the possibilities and limitations of a new method for tuning the superconducting properties of bicrystal Josephson junctions based on  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) thin films. The incorporation of Au nanoparticles changes the growth conditions of the YBCO film in the grain boundary region so that critical current density and normal resistance of such Josephson junctions can be influenced on the same substrate in a different way.

## 2. Sample preparation

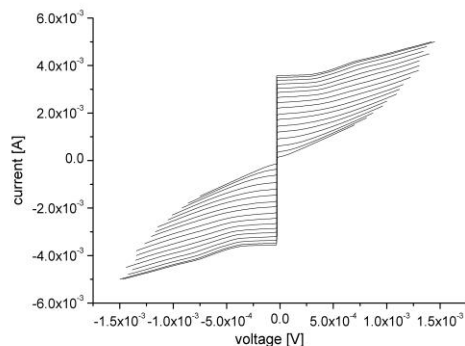
For our investigation we used  $\text{SrTiO}_3$  bicrystal substrates (symmetric and asymmetric grain boundaries) with grain boundary angles of  $24^\circ$ ,  $30^\circ$  and  $36.8^\circ$ . YBCO films with a thickness between 50 nm and 150 nm were epitaxially grown on these substrates by pulsed laser deposition under standard deposition conditions [2]. The superconducting YBCO films in these investigations had a critical temperature of about 90 K, the critical current density without grain boundaries was better than  $3 \cdot 10^6$  A/cm<sup>2</sup> at 77 K. The films were patterned by photolithography and Ar-ion beam etching on a cooled sample holder to avoid degradation in a-b-direction. Using this technique, Josephson junctions and dc-SQUIDs with a junction width between 3  $\mu\text{m}$  and 10  $\mu\text{m}$  across the grain boundaries were prepared.



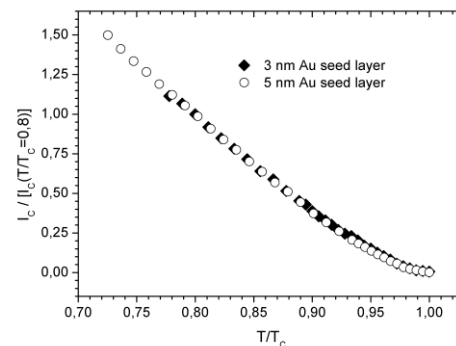
In our investigations we use a very thin sputtered Au seed layer (between 1 nm and 10 nm thickness) which only covers one side of the substrate to ensure a direct comparison between both types of junctions on the same substrate. We found the dewetting of these gold layers to be depending on the seed layer thickness, substrate material, temperature and the vacuum conditions (partial pressure) in the preparation chamber. Thus, by dewetting the seed layer a defined number of Au nanoparticles with a special distribution (size and distance) were formed. During the deposition of the YBCO layer this particle ensemble is modified. Particle diameter, volume distribution and particle density were changed depending on the seed layer thickness as well as on the film thickness of the YBCO layer itself. Meanwhile, the incorporation of self-organized Au nanoparticles also changes the growth conditions of the YBCO thin films [2]. In REM and TEM investigations we found a smoother surface, the suppression of a-axis growth and bending of the YBCO layers in vicinity of the nanoparticles. The result is a small increase of the critical temperature  $T_C$  and a significant increase of the critical current density  $J_C$  at 77 K of the investigated samples with a seed layer thickness between 1 nm and 5 nm. Based on these results we use the same preparation conditions for our bicrystal experiments.

### 3. Experimental results

Near  $T_C$  of the superconducting film we observe a typical foot structure in the resistance-temperature dependence. Depending on the grain boundary angle and the junction width this behaviour can reduce the operation temperature of the Josephson junctions even below the boiling temperature of liquid nitrogen. The current-voltage characteristics of our prepared Josephson junctions with Au nanoparticles show a RSJ like behaviour in a wide temperature range (figure 1). The slope of the extracted temperature dependence of the critical current of Josephson junctions with nanoparticles follows the dependence of junctions without particles [3]. Changes in the seed layer thickness did not affect this behaviour (figure 2).



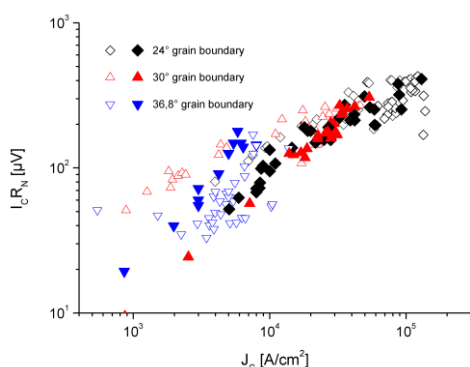
**Figure 1.** Current – voltage characteristics of a Josephson junction ( $30^\circ$  grain boundary) at different temperatures (5 K to 85 K, step width 5 K). The Au seed layer thickness was 3 nm.



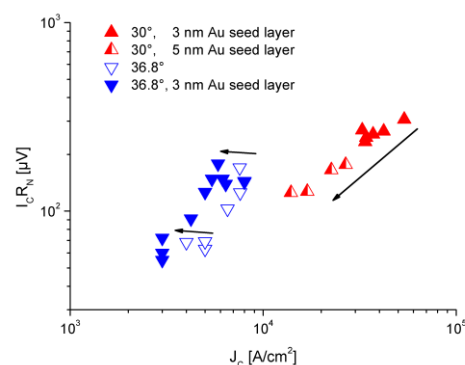
**Figure 2.** Normalized critical current density as a function of normalized temperature for Josephson junctions ( $30^\circ$  grain boundary) with different Au seed layer thicknesses (open symbol 3 nm, filled symbol 5 nm)

In contrast to the experiments with simple microbridges on single crystal substrates, we found a clear reduction of the critical current density [2] of the Josephson junctions in all of the investigated samples for all grain boundary angles. This cannot be explained by a simple reduction of the critical current density by the area or volume of the incorporated normal conducting nanoparticles itself. Furthermore, from REM and TEM investigations we have no indication for any additional agglomeration of nanoparticles along the grain boundary region. The reduction of the critical current density is correlated with an increase of the specific normal resistance of the Josephson junctions. Both dependencies influence the  $I_C R_N$  product of the Josephson junctions. Figure 3 shows an overview over different Josephson junctions and dc-SQUIDS with Au nanoparticles (filled symbols). In the back-

ground we marked former results without nanoparticles (open symbols). In this more or less statistic overview we see that the  $I_C R_N$  product follows the well-known scaling law  $I_C R_N \sim J_C^n$  for bicrystal Josephson junctions with  $n$  close to 0.5 [4]. A more detailed description is only possible if we focus our interest on junctions prepared on the same substrate but with different Au seed layer thickness. This is necessary to minimize the influence of changed preparation conditions during the deposition of different YBCO films on different substrates. Figure 4 shows the same relation for junctions on two different  $36.8^\circ$  bicrystals. The incorporation of nanoparticles from a 3 nm seed layer (filled symbols) creates a reduction of the critical current density but a small increase of the related  $I_C R_N$  product of the Au modified on the same substrate [5]. If we increase the seed layer thickness from 3 nm (filled symbols) to 5 nm (half filled symbols), the reduction of  $J_C$  is stronger in the case of a  $30^\circ$  grain boundary (figure 4). In this case we also found a reduction of the  $I_C R_N$  product on the same substrate.



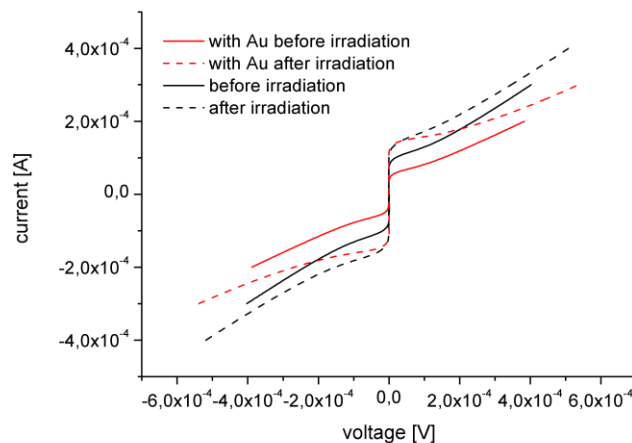
**Figure 3.**  $I_C R_N$  product as a function of  $J_C$  for Josephson junctions and dc SQUIDs with (filled symbols) and without Au nanoparticles (open symbols)



**Figure 4.**  $I_C R_N$  product as a function of  $J_C$  for dc SQUIDs on two  $36.8^\circ$  and one  $30^\circ$  bicrystal substrate. Au seed layer thickness 3 nm (filled symbols) and 5 nm (half-filled symbols). The change in the junction properties on the same substrate is marked by arrows.

The magnetic field dependence of the critical current (77K) for all grain boundary angles shows a more regular, Fraunhofer-like pattern compared to junctions without nanoparticles on the same substrate. A possible leakage current is significantly suppressed. Furthermore, these investigations give no evidence for a more inhomogeneous current distribution inside the Josephson junctions with Au nanoparticles. Another interesting possibility to tune the junction properties is the irradiation with light. Persistent superconductivity can induce additional carriers in oxygen deficient film or grain boundaries, thus increasing the critical current of the Josephson junctions, in the most cases in the order of 10 % or 20 % [7]. The main problem of all these former experiments was the reduction of the normal resistance of the grain boundary junctions during the irradiation. We have done the same experiments for our junctions. A blue light emitting diode (LED, wavelength between 400 nm and 500 nm) was placed near by the junctions inside the magnetically shielded measurement stick. Figure 5 shows the current - voltage characteristics of two junctions with the same junction width on the same bicrystal substrate again. The characteristics were measured before and after the irradiation (30 min) with the LED at 77 K. After the irradiation the sample was warmed up above  $T_C$  and cooled down again to 77 K. In both cases we found an increase of the critical current. But only the Josephson junction with Au nanoparticles also showed a minor increase of the normal resistance and not the reduction measured in all former experiments. In our investigations we found a maximum enhancement of the values of critical current and normal resistance of about 250% and 5%, respectively. Using nanoparticle modified Josephson junctions it was even possible reaching higher critical currents and normal resistances than in non-modified junctions on the same substrate. Correspondingly, a real increase of the  $I_C R_N$  product after irradiation was observed (figure 5). The physics behind this

behaviour is still unclear. The changing growth conditions of the YBCO can create a grain boundary with a lower electron transparency (higher  $R_N$ ) compared to the part without Au nanoparticles. The explanation of the strong increase of the critical current density after irradiation is more complicated. An interaction between radiation and our plasmonically active nanoparticles might be a possible candidate [7]. Additional experiments with different wavelengths should help to solve this question.



**Figure 5.** Current-voltage characteristics for Josephson junctions on the same  $24^\circ$  bicrystal substrate (junction width  $7 \mu\text{m}$ , film thickness  $150 \text{ nm}$ , red curves with Au nanoparticles) before and after irradiation with blue LED.

#### 4. Summary

The incorporation of Au nanoparticles offers the possibility to tune the superconducting properties of Josephson junctions. Critical current density and specific normal resistance can be changed on the same bicrystal substrate depending on the Au seed layer thickness with a local resolution depending on the resolution of the used lithography technology. Irradiation with light can change the junction properties in a more significant way as in the case of unmodified Josephson junctions. Both parameters  $I_C$  and  $R_N$  could be increased in such a manner.

#### 5. Acknowledgment

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#### 6. References

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