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STANFORD CENTER FOR RESEARCH ON  
SUPERCONDUCTIVITY  
AND  
SUPERCONDUCTIVE ELECTRONICS

Technical Report

under

Contract F49620-89-C-0001

for the period

15 October 1989 to 14 October 1990

M.R. Beasley  
Edward L. Ginzton Laboratory  
Stanford University  
Stanford, California 94305

December 1990

STANFORD CENTER FOR RESEARCH ON  
SUPERCONDUCTIVITY  
AND  
SUPERCONDUCTIVE ELECTRONICS

MAJOR ACCOMPLISHMENTS  
(October 1989 to October 1990)

The Stanford Center for Research on Superconductivity and Superconductive Electronics is currently focused on developing techniques for producing increasingly improved films and multilayers of the high-temperature superconductors, studying their physical properties and using these films and multilayers in device physics studies. In general the thin film synthesis work leads the way. Once a given film or multilayer structure can be made reasonably routinely, the emphasis shifts to studying the physical properties and device physics of these structures and on to the next level of film quality or multilayer complexity. The major accomplishments in the past year in all of these areas is summarized in Figs 1 and 2.

Our most advanced thin films synthesis work in the past year has involved developing techniques to deposit a-axis and c-axis YBCO/PBCO superlattices and related structures. Figure 3 highlights some of the central issues involved in this work. More specifically, we have developed a reliable reproducible process for growing films from composite targets in which the composition of the target is preserved in the films and in which the desired structure is formed directly from the vapor—i.e., *in situ* growth. The *in situ* feature is desirable because no solid state reactions with accompanying changes in volume, morphology, etc., that degrade the quality of the film are involved.

Figure 4 illustrates the 90° off-axis geometry. The argon-oxygen sputtering pressure is high enough so that the transport through the plasma is mainly diffusive. The mobility of the species at the surface during the growth is critical so that careful control of the surface temperature is required.

Up until our results on pure a-axis films last December, it was assumed (erroneously) that a-axis films would have rough surfaces. We discovered not only is that incorrect, but that the crystal perfection of a-axis films is superior to c-axis. They grow at lower temperatures, and form better interfaces. All this suggests that a-axis films are excellent candidates for making good sandwich-type Josephson junctions, both of the tunneling and proximity effect varieties. The only good tunnel junctions that have been reported to date on superconductors with  $T_c > 20K$  are the  $BaKBiO_3$  ones which have just recently been reported.

There are no twins in the a-axis films but there are lots of grain boundaries, because the in-plane b and c/3 cell lengths are almost the same. (The resulting "square" in-plane spacing is responsible for the perfection of the interfaces. In contrast, in the c-axis films, the in-plane a and b axes differ by ~2%, resulting in heavy twinning.

The almost perfect square lattice matching between superconducting YBCO and insulating PrBCO makes it possible to grow superlattices, which as judged by the ion channeling experiments, are as perfect as single crystals. Figure 5a, a TEM cross section, shows the superlattice by an out-of-focus image which is sensitive to the electron density. The Pr containing layers are dark. The in-focus image of the atomic rows (Fig. 5b) shows that the structure maintains itself right through the interfaces. The smoothness of the surface is such that no features can be imaged with our most sensitive SEM (resolution ~100Å). The atomic force microscope shows undulations of 40Å over distances thousands of Å. Figure 6 shows the X-ray diffraction scan indicating superlattice satellite peaks, and the RBS channeling data demonstrating high crystalline perfection.



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The grain boundaries between the b and c in-plane orientations in the a-axis films cause excess resistance, as can be seen in the R(T) curves (Fig. 7). Critical currents for a-axis films are reduced by about two orders of magnitude from the c-axis films because of these grain boundaries. This will not degrade vertical transport, however, and this should not be a problem in making junctions.

In collaboration with Xerox, we have also developed techniques for depositing c-axis YBCO films on silicon and silicon-on-sapphire substrates. These films have very good physical properties, although problems remain in growing films thicker than  $\sim 500\text{\AA}$  on the silicon substrates. This work has been summarized by our Xerox colleagues and is not illustrated here.

During the past year, we have also commissioned the new molecular beam synthesis (MBS) system (Fig. 8) that we have been building for several years. Using this system, we have initiated work synthesizing CuO, which involves Cu in the 2+ state as in the high temperature copper oxide superconductors. One important feature of this work is that it augers well for the prospects that we can achieve the synthesis of such highly oxidized states of copper at low vacuum pressures. Our success to date has been made possible by the use of an electron cyclotron resonance (ECR) atomic oxygen beam as our oxygen source during deposition. Figure 9 shows an x-ray diffractometer scan confirming that the monoclinic CuO crystal structure has been achieved and an XPS spectrum demonstrating that the Cu is in the 2+ state. The latter was taken in our MBS system right after the deposition of the film. The epitaxial growth of ultra thin films of CuO should be of great interest.

Using the off-axis sputtering deposition techniques developed earlier to grow high-quality c-axis single layer YBCO films, collaboratively with Hewlett-Packard we have carried out a careful, quantitative study of the role of large-angle grain boundaries in the high residual rf losses universally observed in YBCO thin films. MgO is a good substrate material for microwave devices because it has a low dielectric constant and is cubic. The latter feature insures that the dielectric properties are isotropic in the plane and makes the design of devices simpler. Using YBCO films on

MgO, we have demonstrated that the principal gross defect in these films are large angle  $45^\circ$  grain boundaries as illustrated in Fig. 10. We have previously developed a model of rf losses due to grain boundaries using a Josephson coupled model that is also illustrated in Fig. 10. The idea was to see if these grain boundaries contributed significantly to the measured rf losses, as our model predicted they would. Using in-plane (i.e., in the ab plane) x-ray scans we could not only detect the presence of the large angle grain boundaries but also derive a quantitative measure of their density. In the top of Fig. 11 we show one of these x-ray scans and the area under the scan that arises due to the grains rotated by 45 degrees. In the lower part of the figure we show that the measured surface resistance at 10 GHz increases as the density of such grain boundaries increases. The origin of the remaining residual loss represented by the extrapolation of this curve to zero  $45^\circ$  grain boundaries is not understood. In any event, it is clear from this work that large-angle grain boundaries must be avoided in passive rf applications if the best performance is desired.

Finally, in the past year we have investigated the anisotropy of the Josephson proximity coupling to YBCO thin films. The importance of such studies can be seen from Fig. 12, which shows the various Josephson device structures that are currently being investigated around the world. Some devices couple to YBCO along the c-axis and some along the a-axis. Also remember that the superconducting coherence length is very different along these two directions, the a-axis coherence length being much longer than that along the c-axis. Because of our ability to selectively grow a- or c-axis films, we have been able to compare the coupling to these two directions. The actual device structure fabricated and the corresponding I-V curves are shown in Fig. 13. As is immediately evident from the I-V curves, Josephson proximity coupling to the low temperature superconductor Pb can be achieved on an a-axis film but not on a c-axis film, despite the fact that a state-of-the-art contact resistance was achieved. It is clearly important to establish the reason for this dramatic difference.

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Current Goals:

- Synthesis of increasingly improved HTSC thin films and multilayers.
- Study of their physical properties.
- Device physics studies.

Recent Accomplishments:

- Synthesis of c-axis and a-axis YBCO/PBCO superlattices.
- Successful synthesis of YBCO thin films on Si and Si-on-Sapphire substrates (with Xerox).

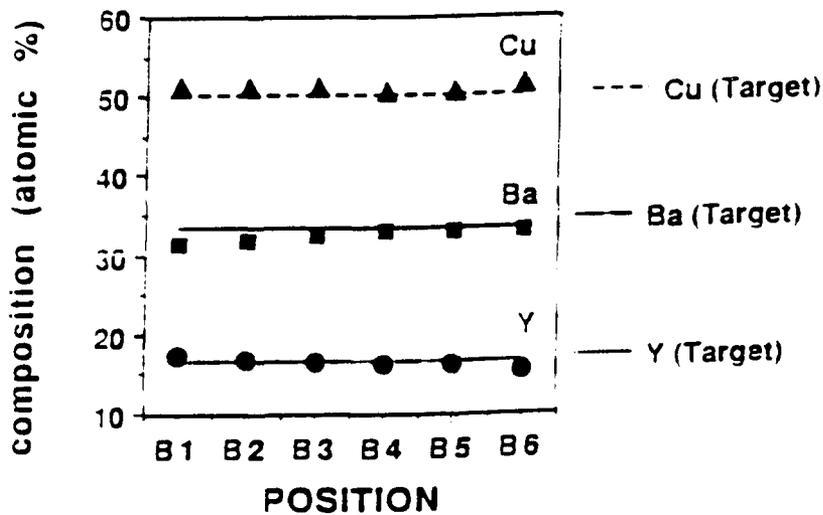
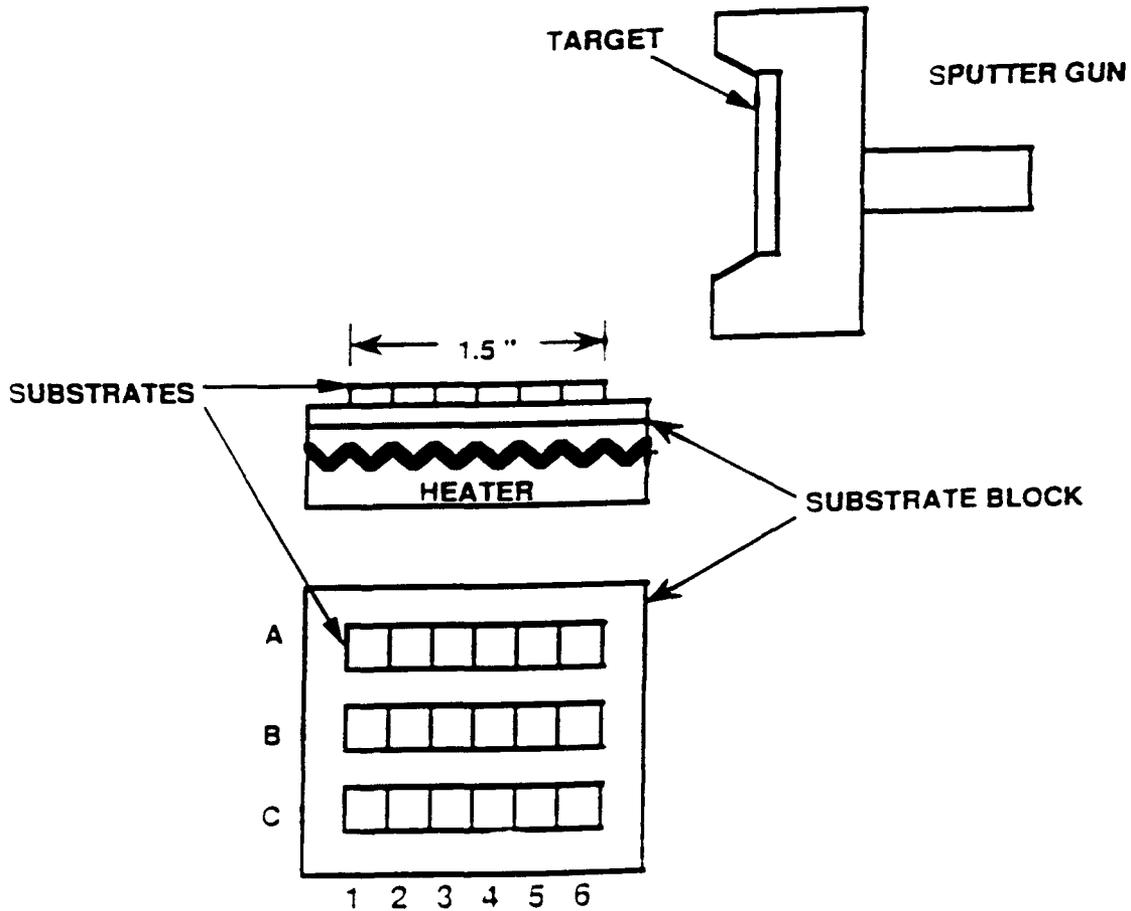
- Commissioning of new Molecular Beam Synthesis (MBS) system—Initiation of studies of pure CuO as model system.
- Demonstration of the importance of 45° large-angle grain boundaries in RF losses (with Hewlett-Packard).
- Comparison of SNS Josephson proximity coupling on a-axis and c-axis YBCO thin films.

# **Fabrication and Electronic Studies of HTSC Thin Films and Multilayers as Prototype Device Structures**

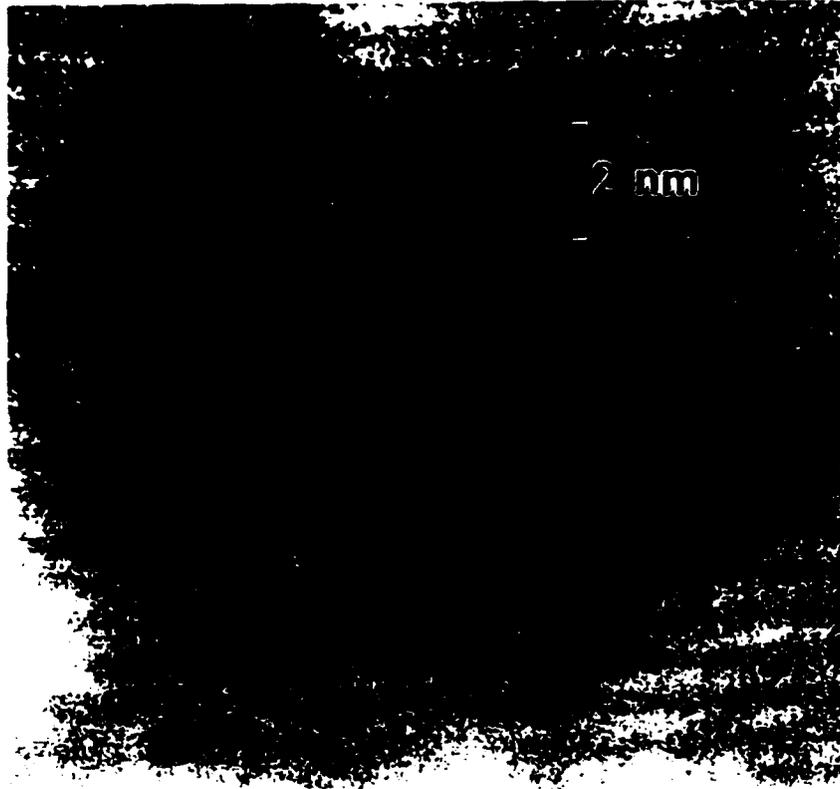
## **Materials Requirements**

- 1. Longer superconducting coherence length  
(a or b direction)**
  - a-axis films and a-axis multilayers**
  - proven by SNS' junctions on a-axis films**
- 2. *In-situ* process for multilayer structures**
  - modified 90° off-axis sputtering**
- 3. Smooth surfaces**
  - sputtered films (atomically smooth)**
- 4. Coherent barrier materials and their growth**
  - PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>**
  - (model experiment YBCO/PBCO superlattice)**
    - \* continuous growth with a-axis**
    - \* perfect lattice match**
    - \* high crystalline quality**

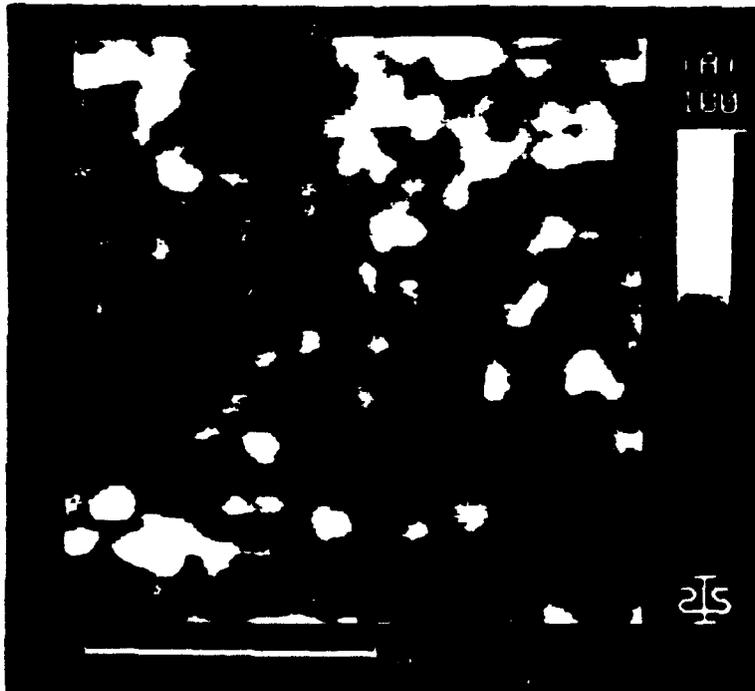
# 90° off-axis sputtering provides large area of uniform composition



Cross-sectional TEM micrograph  
a-axis 12Å YBCO/12Å PBCO Superlattice  
conducting planes  $\perp$  to modulation

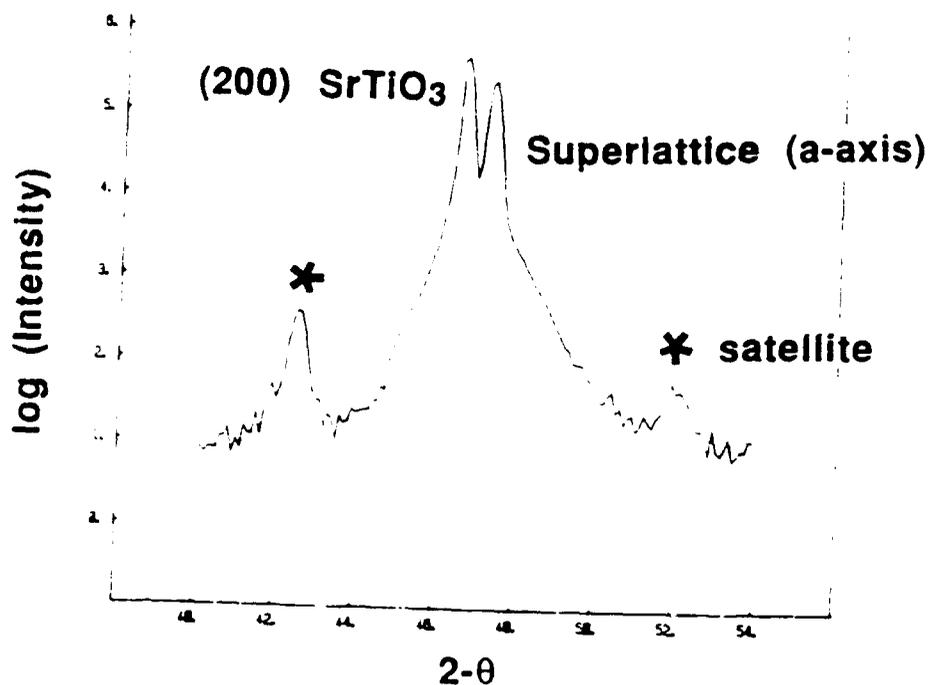


Surface smoothness investigated by Scanning Force  
Microscope. (max feature height  $\sim 40\text{\AA}$ )

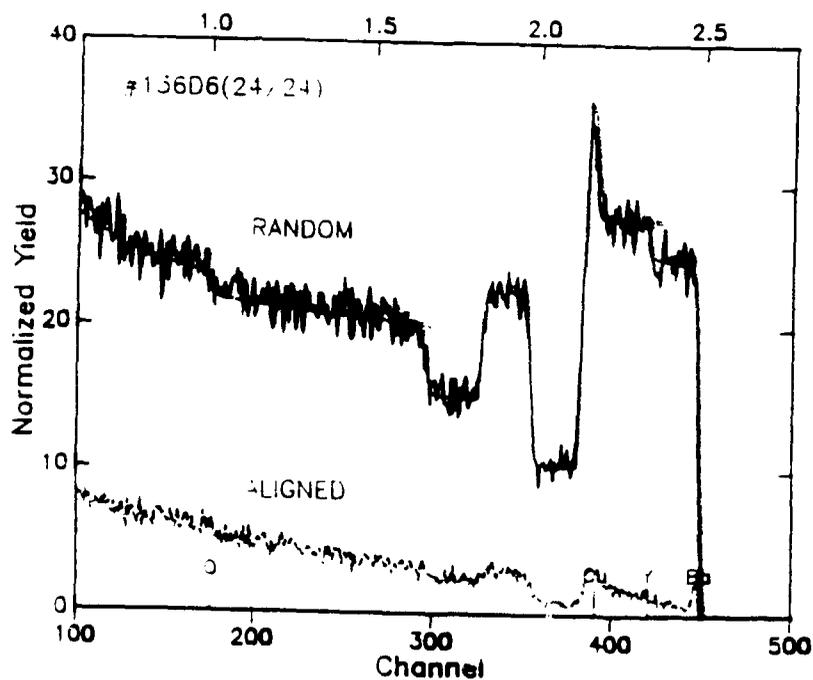


# Crystalline Quality of YBCO/PBCO Superlattices

X-ray diffraction of 12Å/12Å superlattice

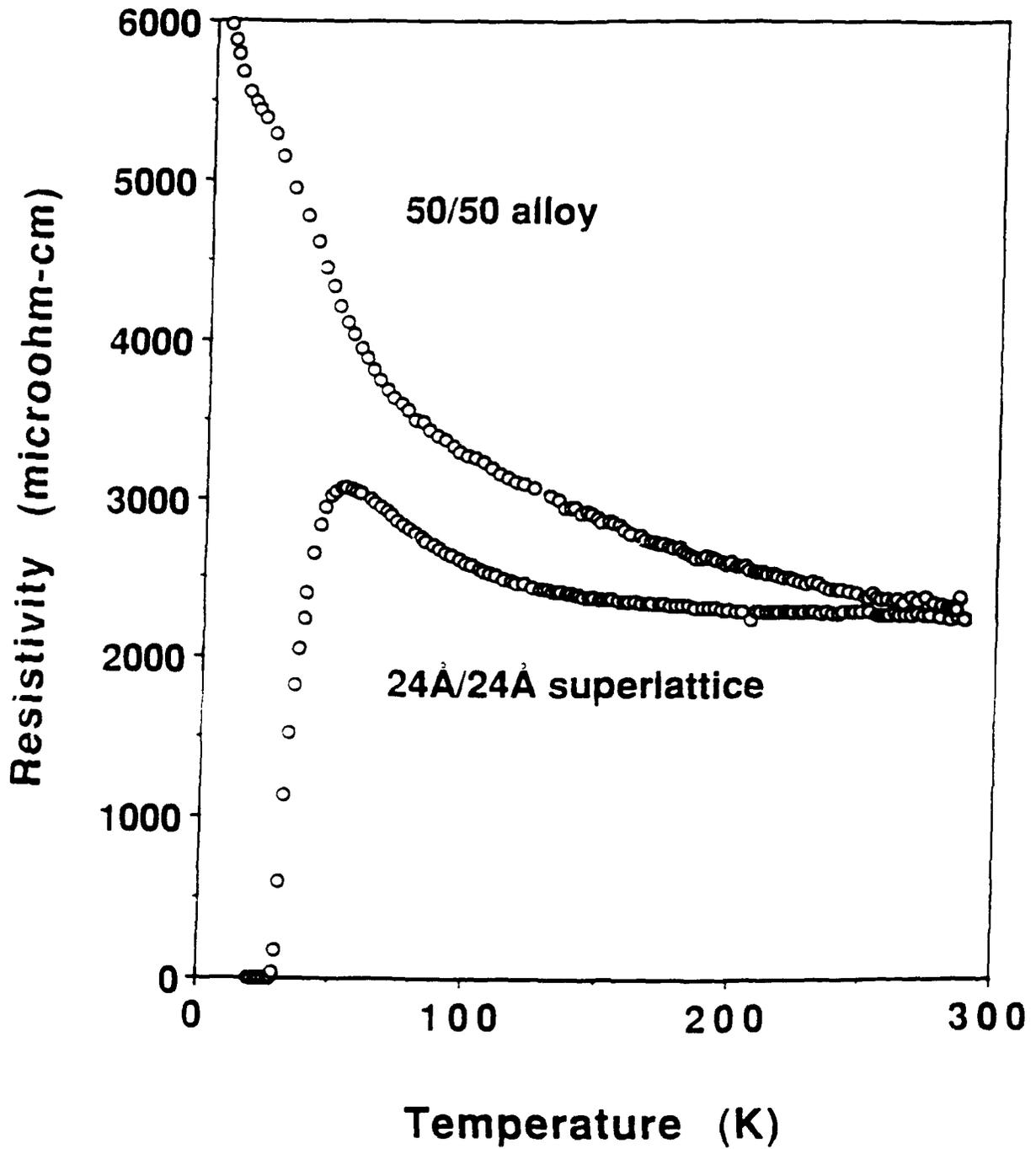


RBS channeling of 24Å/24Å superlattice



$\chi_{\min}(\text{superlattice})=3\%$   
 $\chi_{\min}(\text{single crystal})=3.5\%$

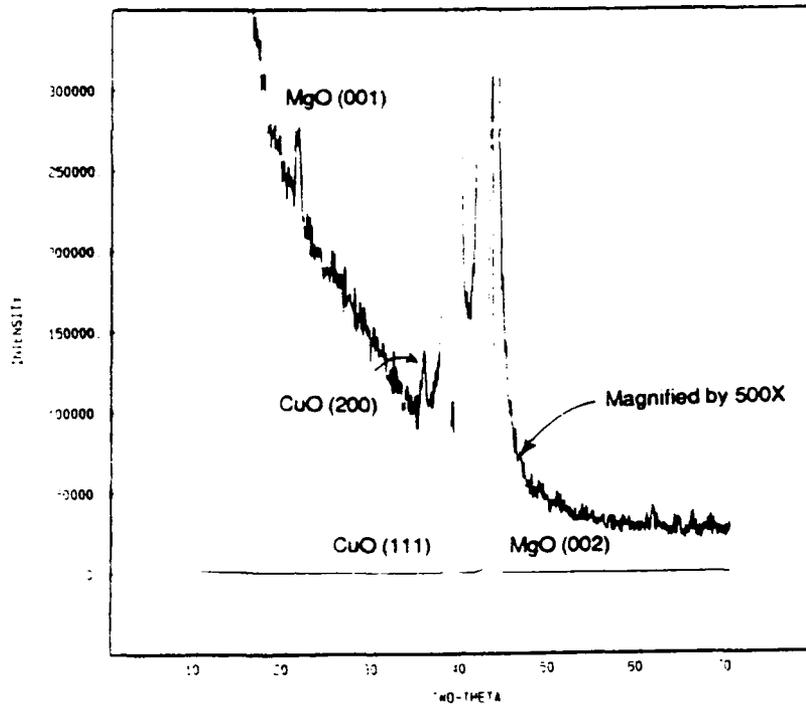
Resistive Transitions of  
YBCO/PBCO a-axis superlattice and alloy  
(long coherence length  $\perp$  surface)



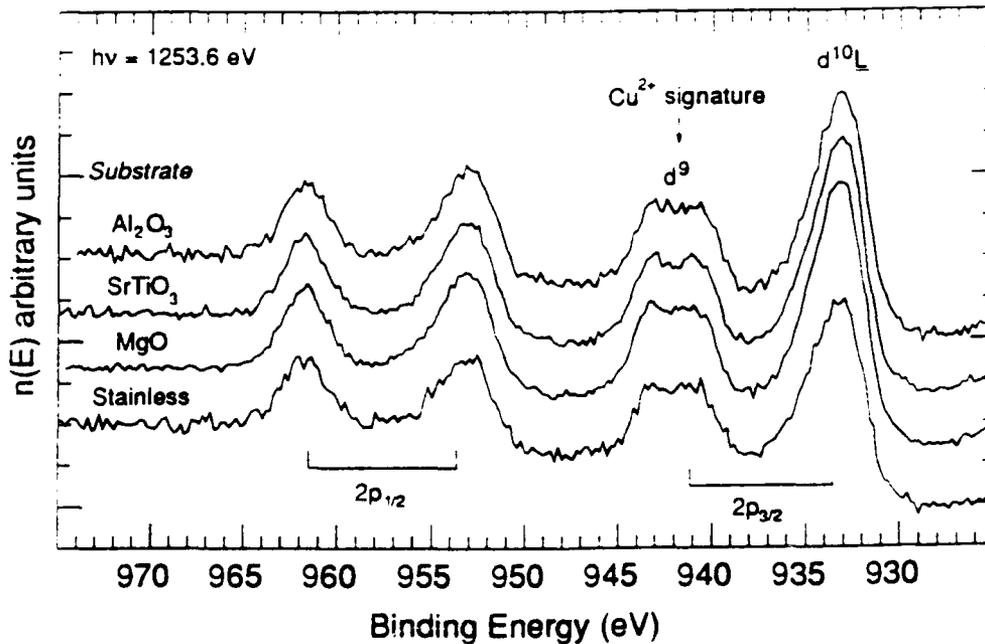


# Evidence of CuO Growth

X-ray diffraction of deposited film

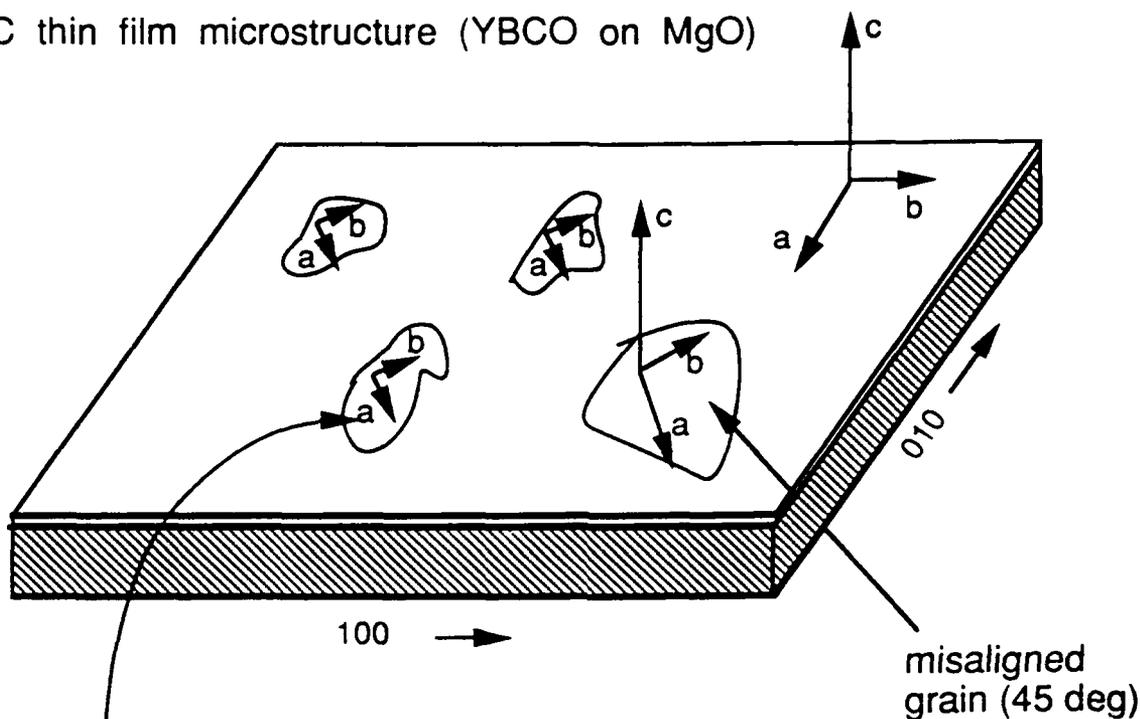


In situ XPS spectrum of film showing characteristic core levels of  $\text{Cu}^{2+}$

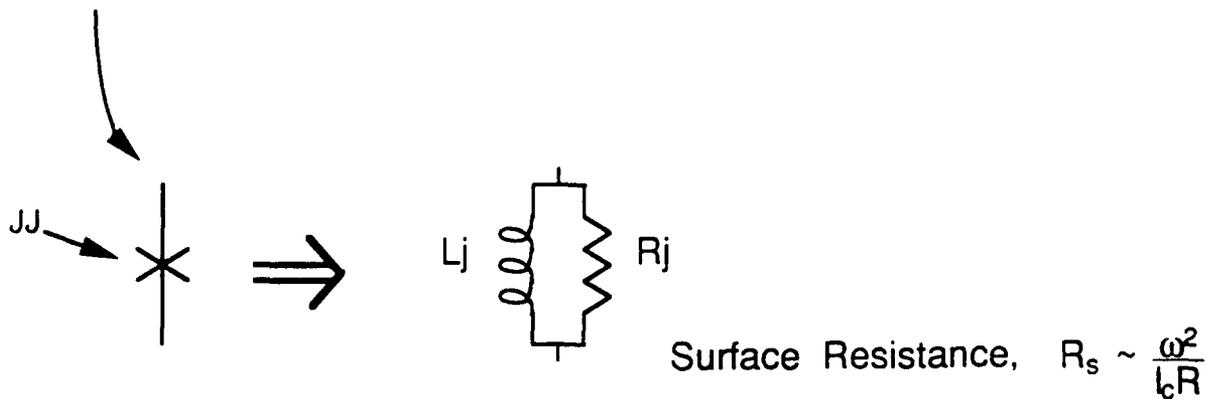


# Effect of Large Angle Grain Boundaries on RF Losses

HTSC thin film microstructure (YBCO on MgO)



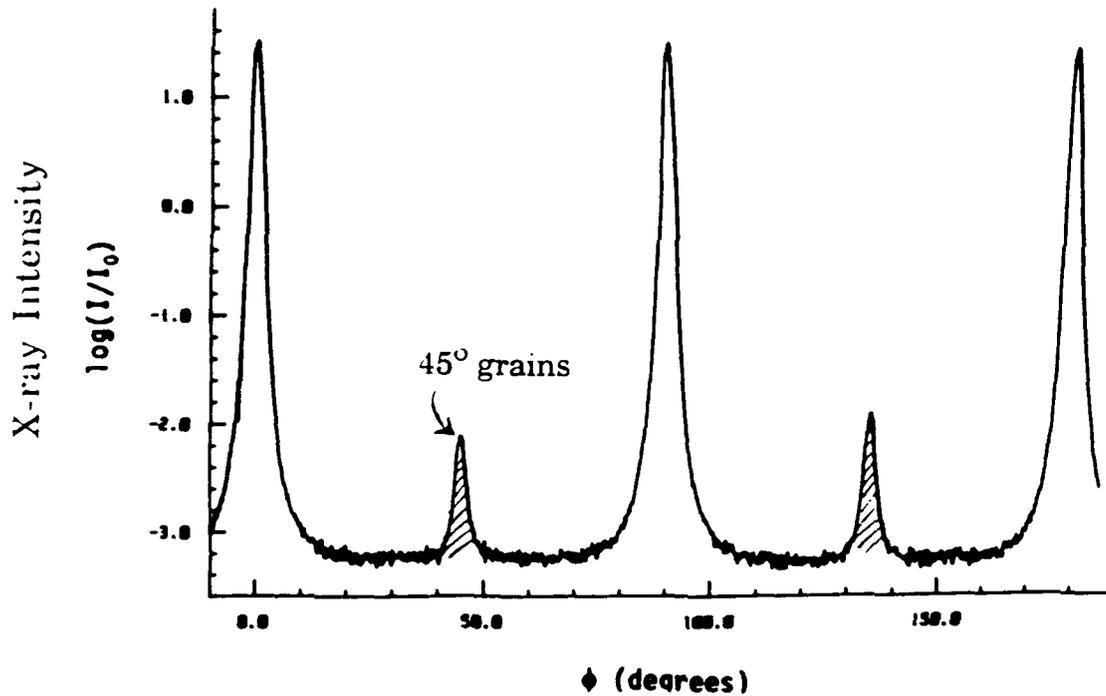
RSJ model for grain boundary weak links



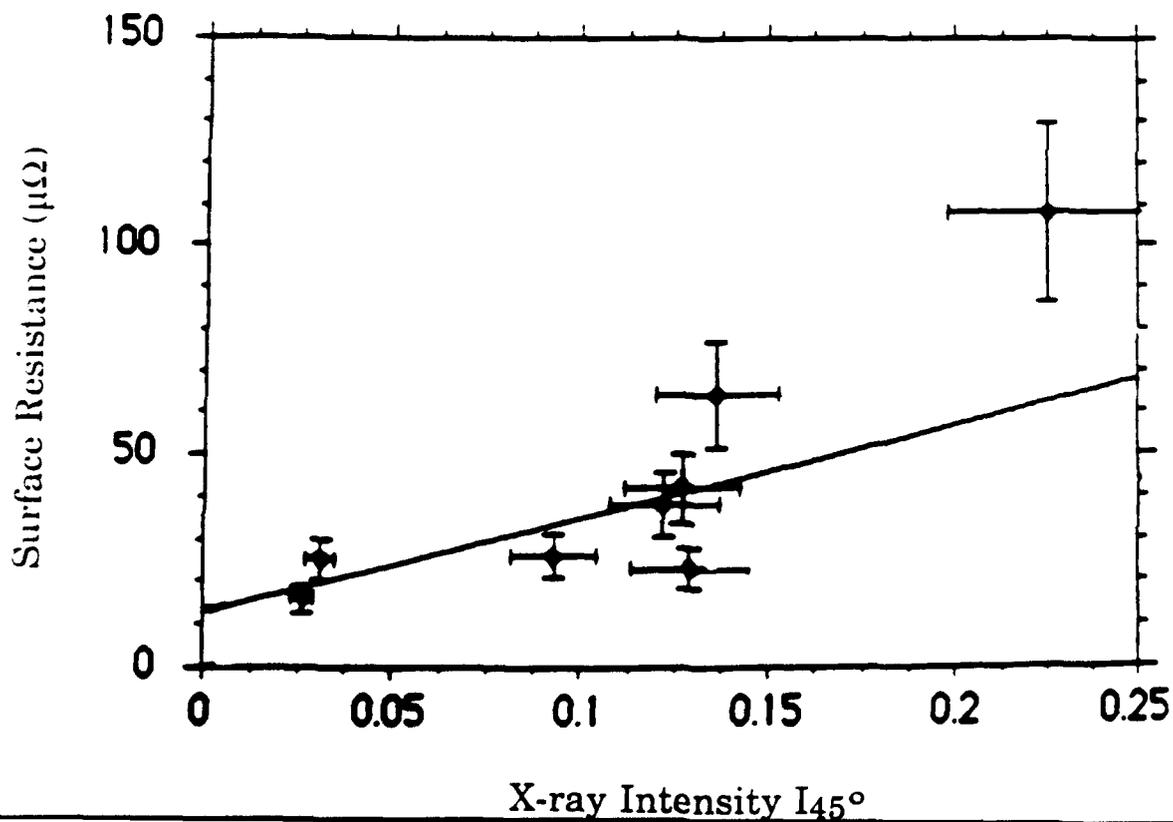
At microwave frequencies these weak links create substantial loss because currents flow through the grain boundaries.

# EXPERIMENTAL CONFIRMATION OF GRAIN BOUNDARY RF LOSSES (WITH H-P)

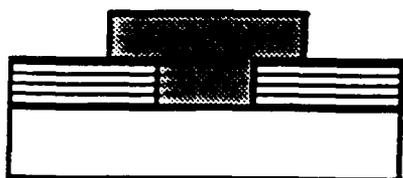
In plane X-ray diffraction → Quantitative measure of # of 45°  
grain boundaries



Measured surface resistance (at 10 GHz) versus grain  
boundary density



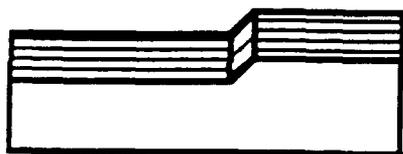
# CURRENT APPROACHES TO JOSEPHSON JUNCTION FABRICATION



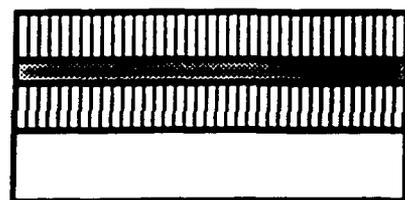
c-axis planar SNS junction



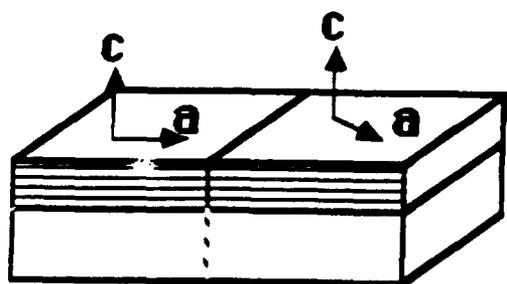
c-axis sandwich SNS junction



c-axis step junction



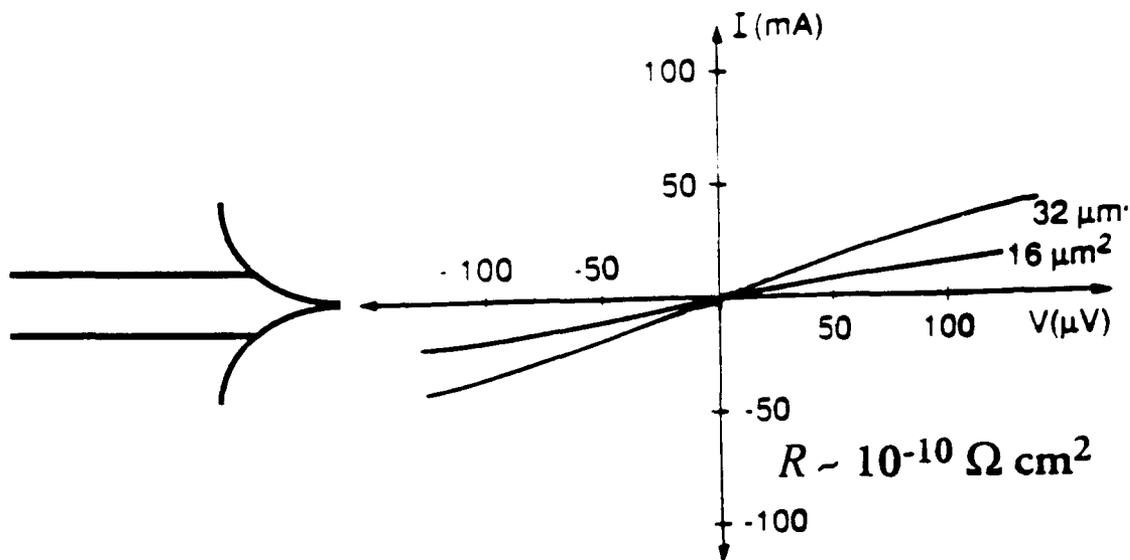
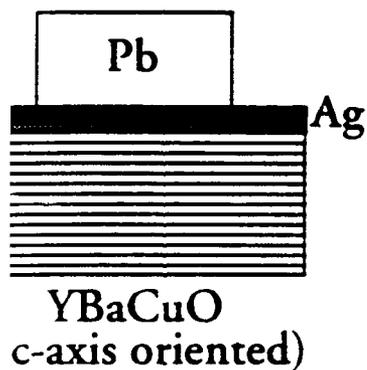
a-axis sandwich SNS junction



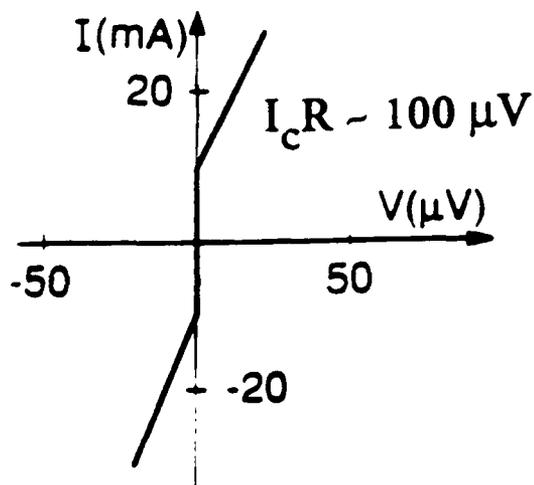
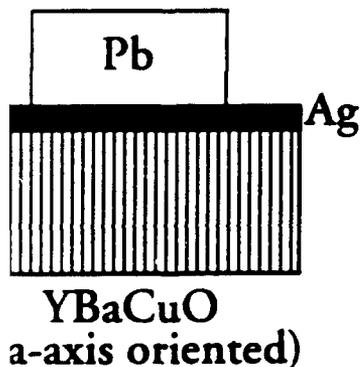
Grain boundary junction

Bicrystal

# Anisotropic Proximity Effect Coupling in YBaCuO-Ag-Pb junctions



No Josephson Coupling



Josephson Coupling