Relation between microstructure and superconducting properties in a-axis 123 films and superlattices

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A-axis oriented superconducting $EuBa_2Cu_3O_7$ films and $EuBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattices have been grown by sputtering on (100) $SrTiO_3$ substrates. Structural analysis by high resolution transmission electron microscopy and X-ray diffraction has been performed. Both kind of samples are well oriented with the c-axis parallel to the substrate plane, the structure at the boundaries is highly coherent and, in the superlattices, high angle X-ray satellite peaks are observed. The presence of $PrBa_2Cu_3O_7$ layers in the superlattices induces a reduction in the superconducting transition temperature that can be attributed to a charge redistribution process in the CuO_2 planes, which in this a-axis orientation belong simultaneously to insulating and superconducting layers.

1. Introduction

The fabrication of anisotropic materials as the high temperature superconductors with different pure orientations is of interest as a complete study of the physical properties can be performed. In the case of a-axis oriented 123 films, the CuO_2 planes are perpendicular to the substrate, so the structural direction with longest coherence length can be analyzed. Also, the preparation of artificial superlattices can allow to control and modify at will the material properties. The a-axis oriented supercoducting/insulating superlattices have been used to reverse the natural anisotropy of 123 compounds [1].

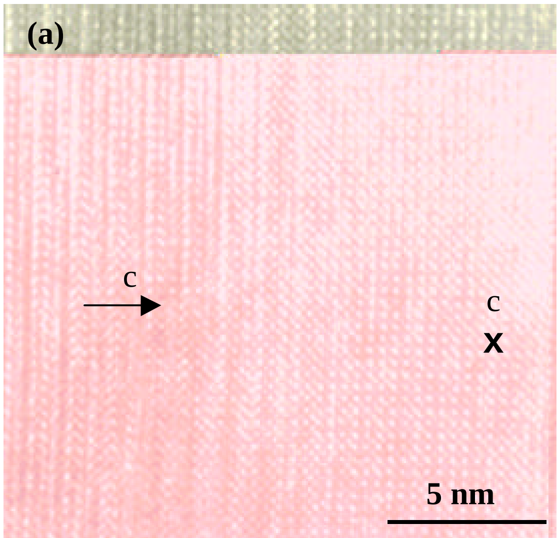
In this work, we have prepared a-axis 123 films and superlattices to study the influence of insulating $PrBa_2Cu_3O_7$ layers on $EuBa_2Cu_3O_7$ superconducting properties in a structure where the CuO_2 planes belong to both materials simultaneously. A careful microstructural characterization has been performed in order to ensure the crystalline orientation of the sample.

2. Experimental

a-axis oriented $EuBa_2Cu_3O_7$ (EBCO) superconducting films and $EuBa_2Cu_3O_7/PrBa_2Cu_3O_7$ (EBCO/PBCO) superlattices have been grown by dc magnetron sputtering on (100) $SrTiO_3$ substrates as reported elswhere [2]. The microstructure of the samples has been characterized by X-ray diffraction (XRD) and high resolution transmission electron microscopy (HRTEM). Specimens suitable for HRTEM were prepared by standard procedures and examined in cross section and in plan view using a Philips CM200 FEG analytical microscope operating at 200 keV and a JEOL 4000 FX at 400 keV. Transport measurements have been performed in order to characterize the superconducting properties of the films and superlattices in a commercial helium cryostat.

3. Results and discussion

The structural characterization by low resolution TEM of the a-axis EBCO samples reveals that they have a pure [100] orientation with the c-axis parallel to the substrate plane, and a very low surface rugosity of less than 5 nm. A closest look into the microstructure can be seen in Fig. 1, where typical HRTEM cross sectional images are shown. In general, cross section is a projection of the structure through the specimen thickness. However, in very thin regions, it is possible to observe that the sample is made up of two different kind of domains, with the c-axis parallel to the substrate plane but either perpendicular or in the plane of the image, separated by 90^0 boundaries (see Fig. 1(a)). In the left part of this image, only the 0.39 nm periodicity corresponding to the b-axis is found whereas in the right part clear fringes separated by 1.17 nm appear, corresponding to the c-axis periodicity. The structure at the 90^0 boundaries is highly coherent, in good agreement with the weak-link free behaviour found in critical current density transport measurements [3].



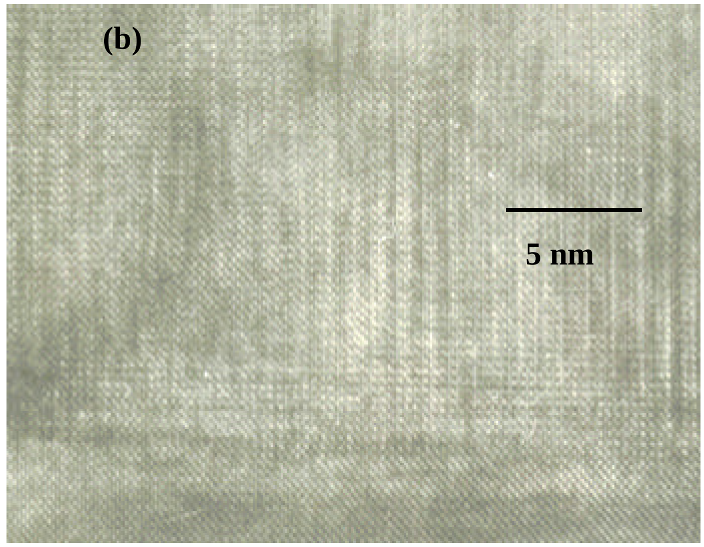


Fig1: (a) Cross sectional HRTEM image of an a-axis film, in a region near the center of the film. Note the two different c-axis directions. (b) Cross sectional HRTEM image of an a-axis film at the substrate-film interface

In order to analyze the initial stages of sample growth, a region close to the film-substrate interface is observed (see Fig. 1(b)). It is found that the initial layer is not continuous. It is formed by small areas of two or three perovskite blocks oriented with the c-axis perpendicular to the substrate surface extended about 20 nm, separated by 4-10 perovskite blocks with the c-axis parallel to the substrate surface. This is an indication of an initial three dimensional growth. Then, after this first stage of nucleation, a-axis oriented growth dominates and only c-axis parallel regions can be observed. This behavior can be associated with the low substrate temperature during deposition that induces a high degree of Y-Ba disorder [4]. Therefore, c-axis oriented growth, that requires the organization of sequential Y and Ba layers, is reduced in comparison with a-axis oriented growth.

The final sample microstructure can be seen in Fig.2, where a plan view image of an a-axis EBCO film is presented. It consists of 20 nm domains separated by 90^0 boundaries, induced by the nucleation along the two principal directions of the cubic $SrTiO_3$ substrate. Therefore, the CuO_2 superconducting planes are configured as sketched in Fig. 2.

Cross-section TEM images of the EBCO/PBCO superlattices also show good growth crystallinity in these samples; continuous fringes perpendicular to the substrate plane separated by 1.17 nm are seen along the whole thickness in a similar way to single films, indicating a coherent growth of both materials in the a-axis direction and that the c-axis is well parallel to the substrate plane. Also a weak contrast between EBCO and PBCO layers is observed in thin areas of the samples in spite of the very similar structural factors. The XRD θ-2θ diagrams reveal the presence of satellite peaks around the principal reflections, corresponding to the superlattice modulation length (see Fig.3(a)). The compositional coherence length (Γ), i.e. the characteristic length where the chemical modulation is maintained, can be estimated from the width of the satellite peaks as Γ = 35 nm and 20 nm for the 16 unit cells EBCO / 5 unit cells. PBCO (16 u.c.EBCO/5 u.c. PBCO) and (7 u.c. EBCO / 6 u.c. PBCO) superlattices respectively. These values correspond to at least 4 bilayers, which is similar to reported values for c-axis superlattices [5]. The main novelty of these well structured a-axis superlattices respect to the c-axis ones is that each CuO_2 plane belongs to EBCO and PBCO layers simultaneously (as it is sketched in Fig. 3(b)), allowing to study the Pr influence on the superconducting planes in a different ordered configuration.

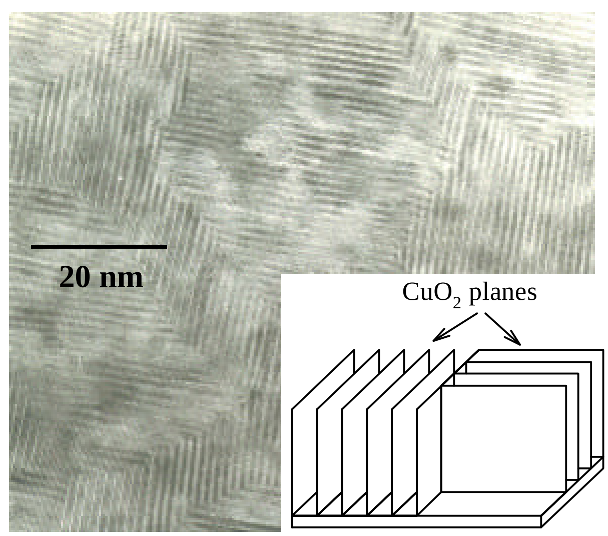


Fig 2: Plan view HRTEM image of an a-axis EBCO film. Inset shows a sketch of the CuO_2 planes indicating the domain microstructure

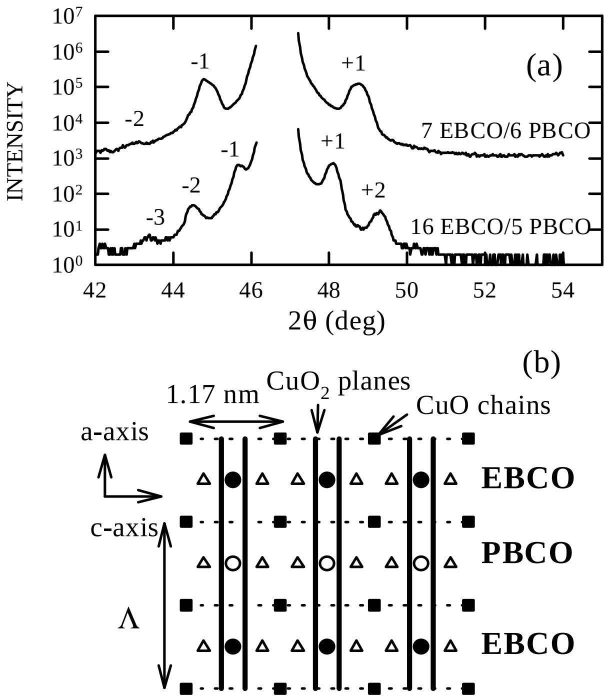


Fig 3: (a) θ-2θ X-ray diffraction scan of a-axis oriented EBCO/PBCO superlattices. (b) Sketch of the atomic microstructure of the a-axis superlattices, showing the superlattice modulation perpendicular to the CuO_2 planes (filled circles, Eu; hollow circles, Pr; triangles, Ba)

The dependence of the superconducting transition on the EBCO and PBCO layer thicknesses is shown in Fig. 4. T_C decreases by reducing the EBCO layer thickness with PBCO layers of only 5 unit cells thick; even in the case of 100 unit cells EBCO layers, this change is important in comparison with a-axis single EBCO films (T_C = 87K), revealing the great influence of Pr atoms on the neighbour CuO_2 planes. In the single $PrBa_2Cu_3O_7$ compound, the presence of Pr supresses superconductivity by reducing or localizing the carriers on the CuO_2 planes [6,7].

Our results indicate that in the a-axis EBCO/PBCO superlattices, the Pr influence extends beyond the insulating PBCO layers into the superconducting EBCO regions, as the CuO_2 planes are shared by both type of layers. This is in agreement with theoretical calculations that predict a charge redistribution in these a-axis superlattices [8].

As the data in Fig. 4(a) reveal, the characteristic length of this phenomenon must extend at least over 40 nm, since already 100 u.c. EBCO layers are affected by the presence of Pr in the superlattice. On the other hand, the superconducting transition is almost independent of the PBCO layer thickness from 5 to 100 unit cells PBCO layers, keeping constant the EBCO layers thickness of 40 unit cells (see Fig. 4(b)).

Therefore, only the first few insulating layers are responsible for the charge redistribution process.

In summary, HRTEM and XRD show that a-axis single EBCO films and EBCO/PBCO superlattices present an ordered structure with CuO_2 planes perpendicular to the substrate and continuous through the whole sample thickness.

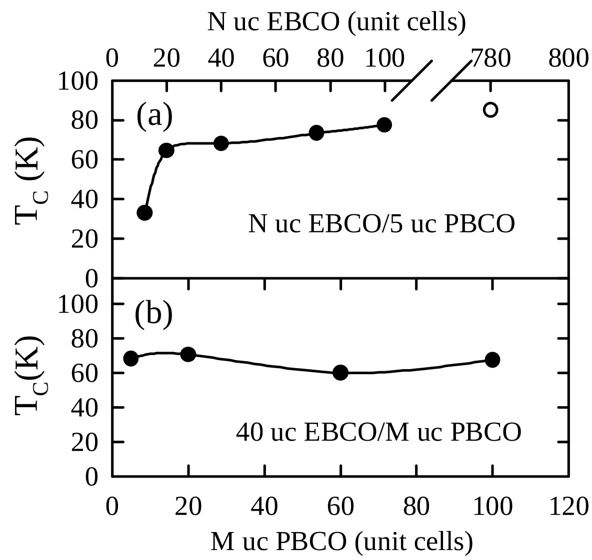


Fig.4: Transition temperature of a-axis EBCO/PBCO superlattices: (a) as a function of EBCO layer thickness (b) as a function of PBCO layer thickness. Hollow circle indicates a single a-axis EBCO film. Solid line is a guide to the eye

In the superlattices, the presence of PBCO layers induces a reduction in the superconducting transition temperature that can be attributed to charge redistribution effects in the CuO_2 planes.

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