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# Superconductivity in BiSrCaCuO Superlattices

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We report resistive measurements on epitaxial superlattices consisting of alternating  $Bi_2Sr_2CaCu_2O_8$  (2212) and  $Bi_2Sr_2CuO_6$  (2201) layers grown by atomic-layerby-layer molecular beam epitaxy (MBE) using oxygen radical beams. The effect of coupling between Cu-O planes was studied by varying the superlattice sequence of different *c*-axis half unit cells. Although depending on the oxygen content, 2201 can be modified from a superconductor to a semiconductor, 2212 one-halfunit-cell-thick layer was found superconducting independing of the conductivity of neighboring 2201 layers. We conclude that all of our superlattices show similar superconductivity in common, indicating the two dimensionality of this cuprate.

### 1. INTRODUCTION

The presence of Cu-O planes in all of the high- $T_c$  superconductors (HTSCs) is believed to be responsible for the strong anisotropy in superconducting properties as well as the basic pairing mechanism. In the Bi<sub>2</sub>Sr<sub>2</sub>Ca<sub>1-1</sub>Cu<sub>1</sub>O<sub>x</sub> (BSCCO) family, the crystal structures differ from one another by the number of consecutive Cu-O planes within a half unit cell. For instance, the  $Bi_2Sr_2CaCu_2O_8$  (2212) phase has pairs of Cu-O planes in successive cells. It is well known that  $T_c$  varies with the number of the Cu-O planes denoted by *l*. In particular, depending on the oxygen content, the Ca-free  $Bi_2Sr_2CuO_6$  (2201) phase with a single Cu-O plane in a half unit cell can be modified from a superconductor ( $T_c \sim 15$  K) to a semiconductor. Although these cuprates are largely anisotropic, it is still unresolved whether an isolated single half-unit-cell layer could exhibit high- $T_c$ superconductivity and what roll the interlayer coupling of the Cu-O planes within a cell and in adjacent cells plays in determining  $T_{\rm c}$ .

In this paper, we report the synthesis and superconducting properties of artificially produced superlattices with different c-axis half-unit-cell sequences. We have picked 2212 and 2201 as an epitaxial growth pair, because their carrier densities are quite similar but they differ in conductivity. Using the atomiclayer-by-layer molecular beam epitaxy (MBE) technique<sup>1.2)</sup> we have been able to tailor the coupling in adjacent 2212 cells by interposing semiconducting 2201 layers.

### 2. SAMPLE PREPARATION

Superlattices consisting of alternating 2212 and 2201 layers have been synthesized in situ by sequentially opening and closing the shutters that control the fluxes from evaporation sources in a versatile MBE system. Oxidation was provided by a flux of atomic oxygen radicals (0<sup>\*</sup>) generated from an rf-excited discharge in a specially designed radical beam source.<sup>1)</sup> The resulting oxygen content of the film is controllable by varying the RF power of the O\* beam source. In this experiment, the MBE growth was carried out under such conditions that the 2201 layer is semiconducting. During growth, the shutters of Bi2O3, Sr, Ca and Cu fluxes were operated by a computer to achieve the c-axis layering of the desired superlattice structure, usually starting with the Bi-O bilayer and ending also with the Bi-O bilayer of 2212. For each element, the shutter opening times were calibrated in order to deposit exactly one monolayer of the corresponding oxide. All the results presented in this paper have been obtained on MgO (100). Typical substrate temperature was 700 ℃. The overall growth rate was 0.5 Å/s with a total film thickness of about 1000 Å.

After the growth, the films were cooled down to room temperature under O\* flux. No further post-deposition annealing was necessary to achieve superconductivity.

It is important to note that there are two distinct in-plane epitaxial relationships<sup>2)</sup> when a thin film of each phase is grown on MgO (100) substrates. Specifically, the a and b axes of the 2212 phase are parallel to MgO [110], while those of the 2201 phase are parallel to MgO [100]. This 45° rotation appears to take place during the Ca deposition in the first sequence of half-unit-cell growth, because the increase of streaking spacing was observed by *in situ* RHEED in this step. Thus, the crystal orientation of the resulting superlattice is rotated by 45° on MgO according as the initial layer is 2212 or 2201.

Figure 1 shows  $\theta - 2\theta$  x-ray diffraction patterns for three different 2212/2201 superlattices. The presence of the well defined satellite peaks on the sides of the main peaks demonstrates that superlattice periodicity was achieved in these structures. The superlattice periods calculated from the separation of two consecutive satellite peaks agreed with ideal



Fig. 1.  $\theta - 2\theta$  x-ray diffraction patterns of three BSCCO/BSCO superlattices, where *m* and *n* denote the half-unit-cell numbers of BSCCO and BSCO, respectively. The arrows indicate superlattice satellite peaks.

values expected from the growth sequences to within a few percent. This confirms that the superlattices are extremely well ordered on a half-unit-cell by half-unit-cell basis. Since BSCCO has no stable intermediate phase in the compositional range between 2212 and 2201, we can rule out the possibility of alloying in contrast with YBaCuO/PrBaCuO superlattices.<sup>3.4)</sup>

### 3. TRANSPORT PROPERTIES

The transport resistivity was measured in a standard four-point geometry using pressed indium contacts. Typical resistive superconducting transitions for several as-grown films showing *c*-axis oriented 2201, 2212, 2223, 2234 and 2245 phases (l = 1-5, respectively) are shown in Fig. 2. In particular, single-phase 2234 and 2245 have not been synthesized by bulk methods, indicating the ability of the atomiclayer-by-layer MBE technique to grow thin films of the desired BSCCO phase. Our data show that apart from 2201,  $T_c$  and the normal-state resistivity vary systematically with the number of Cu-O planes, including 2234 and 2245.

We now discuss the superconducting properties of  $(2212)_m/(2201)_n$  superlattices in which 2212 layers m c-axis half unit cells thick are separated by semiconducting 2201 layers n half unit cells thick. Figure 3 shows the temperature dependence of resistivity for the same superlattices as measured in Fig. 1, i.e.,  $(2212)_1/(2201)_4$ ,  $(2212)_2/(2201)_3$  and  $(2212)_4/(2201)_1$ . Although their  $T_c$  values are



Fig. 2. Typical resistive superconducting transitions for several as-grown films showing  $Bi_2 Sr_2 Ca_{1-1} Cu_1 O_x$  (l = 1-5) single phases.



Fig. 3. Resistive superconducting transitions for the same BSCCO/BSCO superlattices as measured in Fig. 1, where m and n denote the half unit cell numbers of BSCCO and BSCO.

rather different because of the broad transitions, we find that they show similar metallic behavior and that the normal-state resistivity consistently decreases as we increase the number of 2212 half unit cells. These results imply that we are actually observing conduction through the superlattice structures consisting of metallic 2212 and semiconducting 2201 layers that are electrically in parallel. Indeed, the shapes of transitions for (2212)<sub>2</sub>/(2201)<sub>3</sub> and  $(2212)_4/(2201)_1$  are quite similar to that for a typical 2212 single-phase film (as shown in Fig. 1) except for the appearance of a slight resistive tail in (2212)2/(2201)3. Also, in another series of  $(2212)_m/(2201)_2$  samples (m =1, 2 and 4),  $T_c$ , defined as zero resistance, of  $(2212)_4/(2201)_2$  and  $(2212)_2/(2201)_2$  are 59 K and 50 K, respectively. These values are close to T<sub>c</sub> of a typical 2212 film. As for  $(2212)_1/(2201)_2$ , however,  $T_c$  was reduced to around 30 K.

I. Bozovic *et al.*<sup>5)</sup> recently reported that a  $(2212)_1/(2201)_5$  superlattice prepared by MBE using a pure ozone beam exhibited  $T_c \sim$ 75 K, which was essentially equal to  $T_c \sim$  77 K for their 2212 single-phase film. In order to compare this result with ours, it is important to note that their 2201 film is a superconductor showing  $T_c \sim 12$  K. In contrast, our  $(2212)_1/$  $(2201)_4$  sample was prepared under such growth conditions that the 2201 layer is a semiconductor. Thus, the 2212 cells immediately adjacent to 2201 may have their  $T_c$  reduced by 2201. Oxygen deficiency certainly could change the hole concentration and would result in  $T_c$ reduction of the superlattices. Most important, the Cu-O double planes in isolated one-half unit cells of 2212 become superconducting, whether the adjacent 2201 layers show metallic or semiconducting behavior. This would suggest that the coupling between the Cu-O double planes is not essential for superconductivity.

## 4. CONCLUSIONS

We have studied the superconductivity in BSCCO/BSCO superlattices that consist of alternating 2212 and 2201 half unit cells. All of our superlattices showed superconductivity, including superlattices containing isolated Cu-O double planes of 2212 in a semiconducting 2201 matrix. Although their  $T_c$  values are rather different, the overall shapes of superconducting transitions are found quite similar. Most likely, the 2212 cells next to 2201 have their Tc reduced by semiconducting 2201. Also, we cannot see any dependence of  $T_c$  on the separation of the 2212 layers. It seems that the coupling of the Cu-O double planes in adjacent cells is negligible in contradiction to the YBaCu0/PrBaCu0 results.<sup>3,4</sup>) YBaCuO has similar Cu-O double planes but their separation from the next double planes (~ 12 Å ) is shorter than BSCCO (~ 31 Å). The difference of this distance is thought to be responsible for the degree of anisotropy. Thus, we conclude that superconductivity is essentially independent of both the 2212 and 2201 layer thicknesses and BSCCO is a two-dimensional superconductor due to the Cu-O planes within half unit cells.

## REFERENCES

- H. Furukawa, S. Tokunaga and M. Nakao, *Ext.* Abs. 3rd FED Workshop on HiTcSc-ED (1990) p. 72.
- H. Furukawa, S. Tokunaga and M. Nakao, Advances in Superconductivity III, eds. K. Kajimura and H. Hayakawa (Springer-Verlag, Tokyo, 1991) p. 1069.
- J.-M. Triscone, O. Fischer, O. Brunner, L. Antognazza, A. D. Kent and M. G. Karkut, Phys. Rev. Lett. 64 (1990) 804.
- 4) D. H. Lowndes, D. P. Norton and J. D. Budai Phys. Rev. Lett. 65 (1990) 1160.
- 5) I. Bozovic, J. N. Eckstein, M. E. Klausmeier-Brown and G. Virshup, preprint.