Strain and Proximity Effect in $(La,Sr)_2CuO_4$-Based Superconducting Superlattices

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Controlled variations of the in-plane Cu-O bond length and interlayer proximity coupling have been produced in based superlattices $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) based superlattices. In LSCO/Sm_2CuO_4 strained superlattices, the in-plane lattice constant of the LSCO layers is increased and the transition temperature (T_c) is decreased. We estimate the effective pressure to be −8 GPa and conclude the in-plane Cu-O bond length is an important factor controlling T_c. For the LSCO/$La_{1.65}Sr_{0.35}CuO_4$ (metallic) superlattices, a proximity-induced coherence length of 50 Å is calculated for the metallic layer using the de Gennes–Werthamer theory.

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A variety of theories have been proposed to explain the superconducting mechanism in copper oxide superlattices. These are the proximity effect [1,2], carrier concentration [3], pressure effect [4,5], and a 3D-2D crossover (Kosterlitz-Thouless transition) [6]. Hydrostatic pressure studies on bulk oxide superconductors have revealed that the transition temperature (T_c) depends strongly on the pressure [7,8]. However, the use of hydrostatic pressure obscures the intrinsic anisotropy of these materials, since both in-plane lattice spacing a (b) and the out-of-plane c axis are isostatically compressed. Superlattice and multilayer films offer the unique opportunity to separate the roles of the bond lengths within the CuO_2 planes and along the Cu-O apex. In-plane compressive or tensile stress can be easily applied by adjusting the lattice mismatch between the alternate layers [1,9].

In addition, the study of the proximity effect between high-T_c superconductors and normal metals in superlattice structures is both scientifically and technologically of great importance. The coupling between the superconducting and normal-metal layers will help us to better understand the nature and origin of superconductivity in the high-T_c cuprates.

In this paper, we compare the properties of three series of multilayer films containing the superconducting oxide $La_{1.85}Sr_{0.15}CuO_4$. Layers of undoped, semiconducting La_2CuO_4 or Sm_2CuO_4 were inserted in a controlled manner in the LSCO compound using a multitarget, laser-ablation technique to study the effect of in-plane Cu-O bond length variation. Alternatively, layers of overdoped, metallic $La_{1.65}Sr_{0.35}CuO_4$ were inserted to study the proximity effect. Because of the relatively small structural differences between these compounds, complete epitaxy was obtained at the interfaces. Thus systematic trends with the stacking periodicity could be identified, reflecting the changes in the in-plane Cu-O bond length and proximity coupling.

The laser-ablation technique used to form the superlattices has been described in Ref. [9]. The target compositions for the respective layers were $La_{1.85}Sr_{0.15}CuO_4$ (LSCO), Sm_2CuO_4 (SmCO), La_2CuO_4 (s-LCO), and $La_{1.65}Sr_{0.35}CuO_4$ (m-LSCO). The laser beam (pulse intensity: 1 J/cm^2) was sequentially focused on these targets at a repetition rate of 15 Hz to produce an average deposition rate of 0.2 Å/s. Deposition was performed in O_2-O_3(8%) mixtures at a total pressure of 1 mTorr and the substrate was heated at 700 °C. $SrTiO_3$(100) was used as a substrate. The superlattices were formed with stacking periodicities of 1/1 to 120/120 half-unit-cell layers. A periodicity of 1/1 means the periodic stacking of a half unit cell of LSCO and a half unit cell of SmCO (or s-LCO, or m-LSCO). The two layers define one cycle and this cycle was repeated several times. The total thickness of the multilayers was 1000 Å. We have formed three series of superlattices as shown in Fig. 1, namely, $La_{1.85}Sr_{0.15}CuO_4/Sm_2CuO_4$ (LSCO/SmCO; superconductor/semiconductor), $La_{1.85}Sr_{0.15}CuO_4/La_2CuO_4$ (LSCO/s-LCO; superconductor/semiconductor), $La_{1.85}Sr_{0.15}CuO_4/La_{1.65}Sr_{0.35}CuO_4$ (LSCO/m-LSCO; superconductor/normal metal).

Note that in the latter two cases, only the Sr doping level was varied between the successive layers. Since the in-plane lattice parameter a varies only weakly with Sr content over this composition range, negligible interfacial stress was expected for these multilayers. On the other hand, large stress effects were expected for the LSCO/SmCO superlattices, because of the relative large

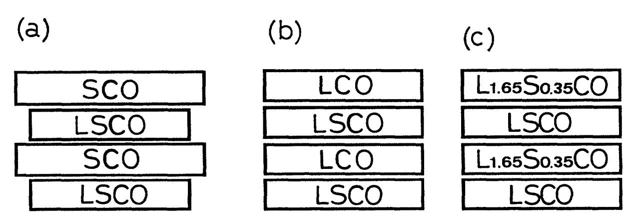

FIG. 1. Schematic models for superlattices. (a) Strained multilayer: $La_{1.85}Sr_{0.15}CuO_4/Sm_2CuO_4$. (b) Superconductor/semiconductor: $La_{1.85}Sr_{0.15}CuO_4/La_2CuO_4$. (c) Superconductor/normal metal: $La_{1.85}Sr_{0.15}CuO_4/La_{1.65}Sr_{0.35}CuO_4$.

lattice mismatch of 3.4% (LSCO, 3.78 Å; SmCO 3.91 Å) for these materials, resulting in an expansion of the *a* axis of the LSCO layers. A necessary condition for the existence of proximity coupling between a superconductor and a normal metal is that the superconducting order parameter extend into the normal metal. The penetration depth of the order parameter depends strongly on the carrier concentration of the normal metal. Thus, different penetration depths were expected for the LSCO/*m*-LSCO and LSCO/*s*-LCO superlattices, respectively, because of the different carrier concentrations. As shown earlier [9], the LSCO/SmCO multilayers can be classified into three categories depending on the stacking periodicity. For the short periodicities, new structures are formed, having a large unit cell containing CuO_5 pyramid, CuO_6 octahedral, and CuO_4 planar Cu-O coordination. At intermediate periodicities, from 10/10 to 60/60, the x-ray diffraction pattern with the scattering vector along with the surface normal (2θ-θ) shows separate peaks corresponding to the *c* axis of both LSCO and SmCO. For these stacking sequences, the in-plane lattice constant *a* is expected to be expanded for LSCO and compressed for SmCO. In order to observe these changes, we have measured the lattice constants *a* and *c* with a four-circle x-ray diffractometer. The results are shown in the form of a pole figure in Fig. 2(a). The figure shows that the LSCO layers and SmCO layers grow epitaxially on the $SrTiO_3$ substrate. The most important observation of Fig. 2(a) is the difference between the two in-plane lattice parameters for LSCO and SmCO. The variation of the in-plane lattice constant *a* with the stacking periodicity is shown in Fig. 2(b). It is seen that the parameter *a* of the SmCO layer decreases with decreasing stacking periodicity below 60/60. On the other hand, for the LSCO layer, *a* increases from 3.78 Å at great thicknesses to 3.84 Å at a periodicity of 15/15 with decreasing layer thickness. As shown in [9], the changes in *c* have the opposite sign of those in *a*. By comparison, for the LSCO/*s*-LCO multilayer, *a* remains constant at 3.84 Å. Thus the expansion of *a* in the LSCO/SmCO layers may be attributed to an in-plane pressure effect due to the large lattice mismatch. The changes in the lattice constant *a* correspond to changes in the in-plane Cu-O bond length in LSCO which is forced to expand parallel to the layer interface as the individual layers become thinner. The pressure at the interface of the strained LSCO layers may be estimated using data for bulk LSCO under isostatic pressure, $dP/da = 1.6\times10^2$ Pa/Å [10]. In the case of the 15/15 multilayer, *a* is expanded from its normal value for LSCO of 3.78 Å to 3.84 Å. The corresponding pressure is 8 GPa.

The pressure effect can also be discussed from another point of view. We consider two sheets in elastic contact and estimate the stress using elasticity theory. In this case, the tensile stress (σ) in the LSCO layer can be written as follows:

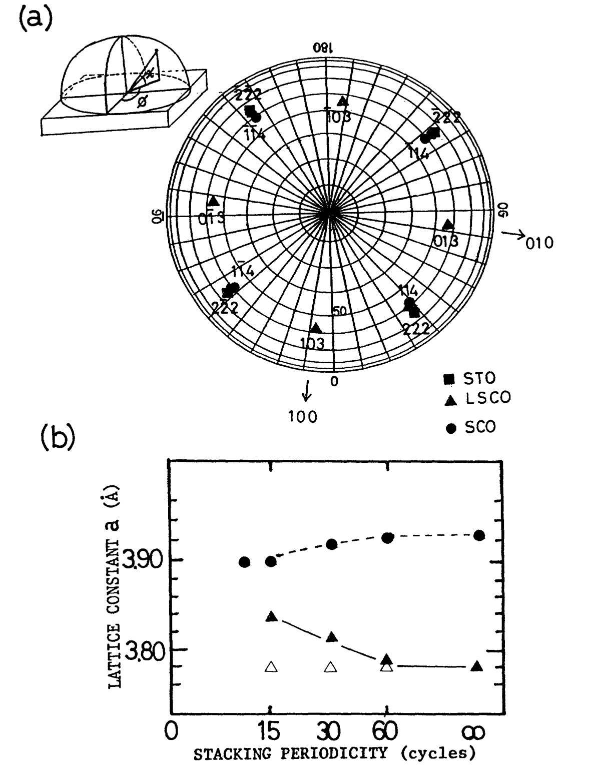

FIG. 2. (a) Pole figure of the LSCO/SmCO superlattice obtained by four-circle x-ray diffraction. (b) Variation of the in-plane lattice constant *a* of the LSCO layer (▲) and SmCO layers (●); △ is in-plane lattice constant *a* of LSCO layer in LSCO/*s*-LCO unstrained superlattices.

$$\sigma = E_i(\alpha_1 - \alpha_2)dT\, dl/t_i\,, \qquad (1)$$

where dl, E_i, α_i, and t_i are the lattice mismatch, Young's modulus, thermal expansion coefficient, and thickness of each layer [$i=1$(LSCO),2(SmCO)]. Using the values of $dT=700$ K and $E_i=1.5$ GPa, and $\alpha=10^{-5}$ K^{-1} [11], the stress in the LSCO layer can be estimated as 3.0 GPa. This value is similar to the pressure derived above from the experimentally observed changes in lattice parameter.

Corresponding to the changes of the lattice constant, the critical temperature (T_c) of these strained superlattices decreases rapidly with decreasing number of half unit cells as shown in Fig. 3(a) (squares and solid line). It should be noted that the in-plane pressure effect is independently induced in these superlattices. Using the value -8 GPa, a negative pressure effect of -2.0 K/GPa can be estimated from the observed T_c decrease of Fig. 3(a).

For the unstrained superlattices, LSCO/*s*-LCO and LSCO/*m*-LSCO, on the other hand, the lattice constant *a* remains constant over the entire range of layer thicknesses. For the former, T_c does not change down to a

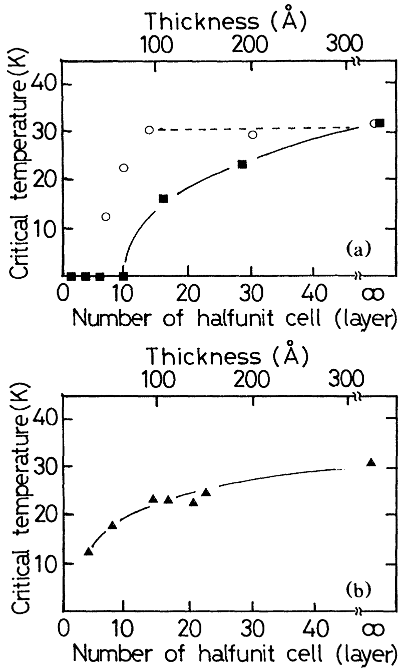

FIG. 3. Variation of the critical temperature T_c of (a) LSCO/SmCO (solid line), LSCO/LCO (super/semiconductor, dashed line), and (b) $LSCO/La_{1.65}Sr_{0.35}CO_4$ (super/normal) multilayer films as a function of the number of half unit cells.

periodicity of 15/15 multilayers. The T_c reduction for layer thicknesses less than 10/10 periodicity will be discussed later.

The variation of T_c for the super/normal superlattices is shown in Fig. 3(b). T_c decreases gradually with decreasing number of half unit cells; however, the T_c decrease deviates markedly from that of the strained multilayers. In this system, there is essentially no strain at the interface of each layer. Therefore, the question is why the T_c of this kind of superlattice decreases with decreasing stacking periodicity in the absence of lattice mismatch.

There are several possible explanations for this tendency of T_c, such as interlayer coupling, 3D-2D (Kosterlitz-Thouless) transition, and proximity effect.

In the following we will examine the possibility of proximity coupling in greater detail. It is well known that the Cooper pairs penetrate from a superconductor into a normal metal, producing a lowering of T_c in the superconducting material [2,3]. In the case that each layer is relatively thick, we calculate T_c using the de Gennes–Werthamer (dGW) theory [12,13]. For thin layers, the coupling is calculated according to the Cooper limit theory [14].

The relation between T_c and the thickness of each layer may be written as follows [13]:

$$\ln(T_{c_s}/T_c) = X(\xi_s^2 k_s^2) , \tag{2}$$

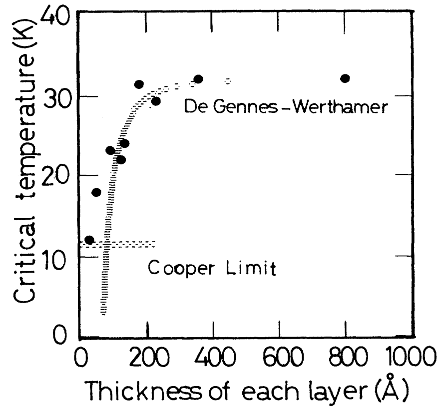

FIG. 4. Critical temperature of the LSCO/SmCO superlattices as a function of the thickness of each stacking layer. Solid circles are the measured values and open circles are calculated using the dGW theory and the Cooper limit theory, respectively.

$$[N\xi^2 k \tan(kd)]_s = [N\xi^2 \tanh(kd)]_n , \tag{3}$$

$$X(z) = \Psi(\tfrac{1}{2} + \tfrac{1}{2} z) - \Psi(\tfrac{1}{2}) , \tag{4}$$

where Ψ is the digamma function and T_{c_s}, T_c, k, and d_i are the T_c of superconductor, T_c of normal metal, Boltzmann constant, and thickness of each layer, respectively.

The open circles in Fig. 4 represent the calculated value using the dGW theory, using the parameters $V_F = 8\times 10^6$ cm sec^{-1} and $T_c = 34$ K [15]. It is seen that for $d > 150$ Å, the dGW theory is in good agreement with the observed data (solid circles). However, below 150 Å, the measured T_c is significantly higher than that predicted by the dGW model. Instead, the observed values approach the value of the Cooper limit calculated using the average $(N_0 V)$ of the super/normal system given by

$$(N_0 V) = (N_s^2 V_s d_s + N_n^2 V_n d_n)/(N_s d_s + N_n d_n) \tag{5}$$

and using

$$T_c = \Theta_D / 1.45 \exp(-1/N_0 V) , \tag{6}$$

where Θ_D, $N_i V_i$, and d_i are the Debye temperature, the effective attractive interaction, and the thickness of each layer [$i = s$(super),n(normal)], respectively [14]. This limit simply corresponds to very thin layers where the electrons experience the average pairing interaction of the n (normal) and s (super) materials.

By fitting Eqs. (2)–(4) to the experimental data, a coherence length of 50 Å is estimated for the normal m-LSCO layers. According to the BCS theory, the coherence length of the oxide superconductors is very short because of their high superconducting critical temperature (T_c). Therefore weak interaction between electrons and phonons in the normal layer produces this large coherence length of 50 Å. This may be one possible explanation for the long proximity coupling length.

In conclusion, we have formed three series of multilayers, LSCO/SmCO, LSCO/*s*-LCO, and LSCO/*m*-LSCO by using a multitarget pulsed-laser-ablation technique. In the case of LSCO/SmCO strained multilayers, the lattice constants a and c of the LSCO layers are expanded and contracted, respectively. In this superlattice, a pressure of 8 GPa at the interface can be estimated by comparing with isostatic pressure data on the bulk system. A value of 3 GPa is estimated from the thermal expansion elastic calculation. This result shows that uniaxial pressure, along the basal plane, giving rise to variations in the Cu-O bond length in the CuO_2 plane is one of the important factors determining T_c. In the case of the unstrained superlattices of LSCO/*s*-LCO, on the other hand, the lattice constants a and c do not depend on the layering sequence. The T_c decrease in the unstrained superlattices of superconducting/overdoped-*m*-LSCO layers was discussed in terms of proximity coupling, and a large coherence length of 50 Å was estimated for the normal-metal layer.

[1] J.-M. Triscone, D. Ariosa, M. G. Karkut, and O. Fischer, Phys. Rev. B **35**, 3238 (1987).
[2] W. P. Lowe and T. H. Geballe, Phys. Rev. B **29**, 4961 (1984).
[3] T. Nishino, E. Yamada, and U. Kawabe, Phys. Rev. B **33**, 2042 (1986).
[4] A. Gupta, R. Gross, E. Olsson, A. Segmuller, G. Koren, and C. C. Tsuei, Phys. Rev. Lett. **64**, 3191 (1990).
[5] K. Horiuchi, M. Kanai, T. Kawai, and S. Kawai, Phys. Rev. B **45**, 13 152 (1991).
[6] M. Rasolt, T. Edis, and Z. Tesanovic, Phys. Rev. Lett. **66**, 2927 (1991).
[7] R. Griessen, Phys. Rev. B **36**, 5284 (1987).
[8] Y. Yamada, T. Matsumoto, Y. Kaieda, and N. Mori, Jpn. J. Appl. Phys. **29**, 1250 (1990).
[9] H. Tabata, T. Kawai, and S. Kawai, Appl. Phys. Lett. **58**, 1443 (1991).
[10] S. Yomo, C. Murayama, H. Takahashi, N. Mori, K. Kishio, K. Kitazawa, and K. Fueki, Jpn. J. Appl. Phys. **26**, L603 (1987).
[11] X.-D. Xiang, J. W. Brill, L. E. Delong, L. C. Bourne, A. Zettl, J. C. Jones, and L. A. Rice, Solid State Commun. **65**, 1073 (1988).
[12] P. G. de Gennes, Rev. Mod. Phys. **36**, 225 (1964).
[13] N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).
[14] L. N. Cooper, Phys. Rev. Lett. **6**, 689 (1961).
[15] S. A. Wolf and V. Z. Kresin, in *Proceedings of the International Symposium on Superductivity*, edited by T. Ishiguro and K. Kajiimura (Springer-Verlag, Tokyo, 1989), p. 447.