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# Weak Ferromagnetism of $La_{1.99}Sr_{0.01}CuO_4$ Thin Films: Epitaxial Strain and Corrugation in $CuO_2$ planes

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The weak ferromagnetism of $La_{1.99}Sr_{0.01}CuO_4$ epitaxial thin films is investigated using magnetoresistance measurement. While a step-like negative magnetoresistance associated with the weak ferromagnetic transition is clearly observed in the films grown on $YAlO_3$(001), it is notably suppressed in the films grown on $SrTiO_3$(100) and $(LaAlO_3)_{0.3}(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$(100), and almost disappears in films grown on $LaSrAlO_4$(001). The strong suppression of the step-like magnetoresistance provides evidence that the $CuO_2$ planes are much less corrugated in thin films grown on tetragonal substrates, particularly on $LaSrAlO_4$(001), than in bulk crystals.
Key words: LSCO, weak ferromagnetism, spin canting, epitaxial strain

## 1. INTRODUCTION

Epitaxial growth of $La_{2-x}Sr_xCuO_4$ (LSCO) superconductors has been gathering much attention because of its large effects on superconducting properties [1-3]. In these studies, in-plane compressive and out-of-plane tensile lattice strains were reported to be favorable to higher $T_c$. As is widely known, a bulk crystal of LSCO has two phases: high-temperature tetragonal (HTT, *I4/mmm*) and low-temperature orthorhombic (LTO, *Bmab*) phases. In the LTO phase, a staggered rotation of the $CuO_6$ octahedra gives rise to a characteristic corrugation in the $CuO_2$ plane. Considering that superconductivity occurs in this LTO-phase stable region for bulk crystals, we first need to know how the epitaxial strain influences this corrugated structure. However, the neutron diffraction, which is the most powerful (and probably the only) technique for studying the structure of bulk crystals, is considered to be ineffectual for this particular purpose, because the volume of thin-film sample is too small. Thus far, the structure of $CuO_2$ plane of the strained films has remained as an open question.

An indirect but prospective approach to the corrugation in the $CuO_2$ is to study an antiferromagnetic sample instead of a superconducting sample. It was well known that the antiferromagnetic long-range order (AF-LRO) of LTO-LSCO ($x<0.02$) has close relation to the corrugation of the $CuO_2$ plane. In this AF-LRO state, spins on $Cu^{2+}$ ions show characteristic canting according to the buckling of the $CuO_6$ octahedra. Due to this canting, antiferromagnetic LSCO shows metamagnetic transition when magnetic field is applied perpendicular to the $CuO_2$ plane, which has been known as a weak-ferromagnetic (WF) transition. The WF transition strongly affects the charge transport as was first suggested by Thio *et al.* [4]. Recently, Ando *et al.* have discovered that the in-plane magnetoresistance also couples with the WF transition [5], where the in-plane magnetoresistance exhibits steep decrease across the WF transition as shown in Fig.1. Therefore, the in-plane magnetoresistance can be used as a sensitive probe of the weak ferromagnetism, which is a good measure of the corrugation in the $CuO_2$ plane. In this paper, we show the results of magnetoresistance measurement for $La_{1.99}Sr_{0.01}CuO_4$ epitaxial thin films to discuss the corrugation of the $CuO_2$ plane [6].

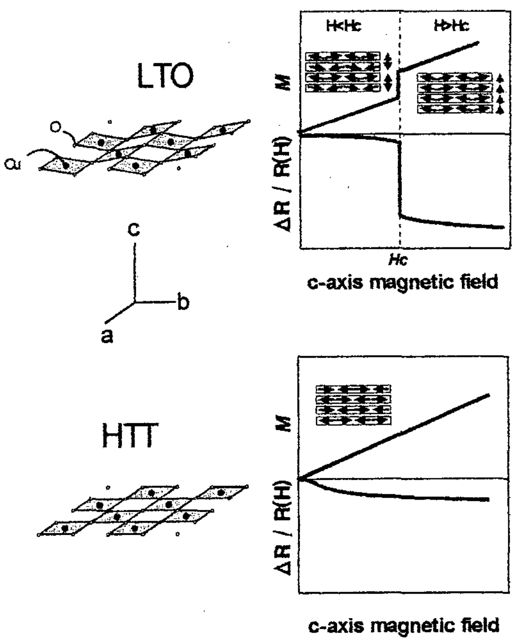


Fig.1 Concept of field dependence of magnetization and in-plane resistivity. Discontinuous jump appears in LTO-LSCO but should not appear in HTT-LSCO.

## 2. EXPERIMANTAL RESULTS

### 2.1 Sample preparation

$La_{1.99}Sr_{0.01}CuO_4$ thin films were prepared by pulsed-laser deposition (KrF excimer, $\lambda$ = 248 nm). The details will be reported elsewhere [6]. At this Sr concentration, we expect that the AF long-range order appears at $T \approx 200$K in a bulk crystal [7]. We used four substrates: orthorhombic $YAlO_3(001)$ has a rectangular surface symmetry, while tetragonal $LaSrAlO_4(001)$, cubic $(LaAlO_3)_{0.3}(SrTa_{0.5}Al_{0.5}O_3)_{0.7}(100)$, and cubic $SrTiO_3(100)$ have a square surface symmetry. Hereafter, these substrates are referred to as YAP, LSAO, LSAT, and STO, respectively. To avoid confusion, we follow the axis notation of the LTO structure of LSCO. Film thickness was set at approximately 1800Å. After patterning the films for four-terminal measurements, they were carefully annealed at 600℃ in helium to remove extra oxygen following the procedure for lightly doped bulk crystals [7]. Since the extra oxygen can easily induce superconductivity, as discussed by Sato *et al.* [2] and Bozovic *et al.* [8], this post annealing in helium is crucial for the present experiments.

2.2 X-ray diffraction

All of the films are highly *c*-axis oriented. The *c*-axis length estimated from the observed 00*l* (*l* : even) reflections is summarized in Fig.2 as a function of the lattice parameter of substrates with the data of ceramic samples [9]. Although the *c*-axis length does not show monotonic behavior at first glance, this variation is consistent with the results reported by Sato *et al.* for $La_{1.85}Sr_{0.15}CuO_4$ films [1], and shows that a significant change in the *c*-axis is observed only in the films with sufficiently small lattice misfit to the substrate.

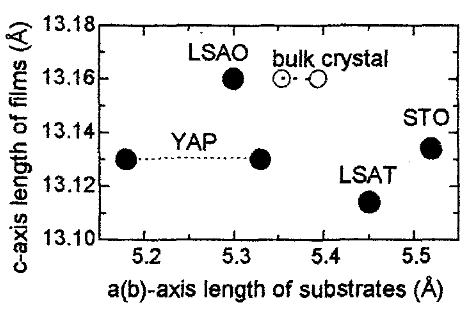


Fig.2 The *c*-axis length of the films as a function of the *a*(*b*)-axis length of the substrates.

In-plane crystallographic symmetry was confirmed by a pole-figure goniometer to scan both the 208 and 028 reflections (Fig.3(a)). Figures 3(b)-3(e) show the single scans of the 208 and 028 reflections. For the film grown on YAP, the 208 and 028 reflections appear at different angles indicating an orthorhombic symmetry. The in-plane orientation is determined as YAP [100] ‖ LSCO [100], which may allow us to expect that the corrugation survives in the $CuO_2$ plane. On the other hand, the two reflections appear at the same angle for the films grown on LSAO, LSAT, and STO. However, these results do not immediately mean that these films are really tetragonal. For example, $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$ is orthorhombic and does not become tetragonal even when grown on STO(100), but it exhibits a twin structure composed of orthorhombic domains [10]. Thus, we cannot judge whether the corrugation is actually removed from these three films at this stage, and therefore, the magnetoresistance measurement becomes important.

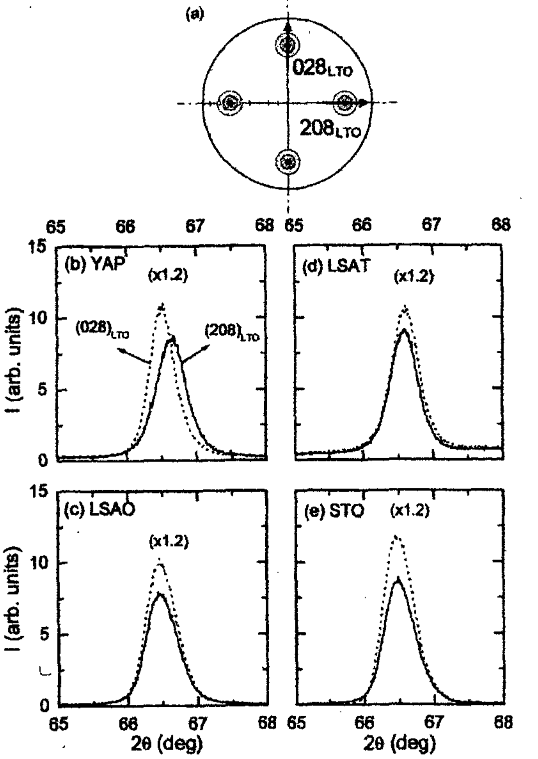


Fig.3 (a) Scan direction of 208 and 028 reflections by pole figure goniometer. (b)-(e) The 208 and 028 reflections of the films.

2.3 Resistivity

Figure 4 shows the in-plane resistivity of all films. The films grown under the compressive strain (YAP and LSAO) show a higher resistivity than those grown under the tensile strain (LSAT and STO). The orthorhombic LSCO film grown on YAP shows anisotropic resistivity along the *a* and *b* axes. $\rho_a$ is always lower than $\rho_b$ for this film, and the temperature where the resistivity shows a minimum value is also lower for $\rho_a$ than for $\rho_b$. Such in-plane anisotropy is qualitatively consistent with those for bulk crystals with $x < 0.02$ [11], which supports our identification of the *a*- and *b*-axis directions. The other three films show almost no in-plane anisotropy, which is also consistent with the results of x-ray diffraction.

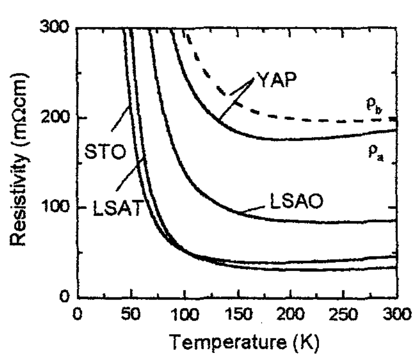


Fig.4 Temperature dependence of the in-plane resistivity.

2.4 Magnetoresistance

After characterizing the films by x-ray diffraction and resistivity measurements, we measured the in-plane magnetoresistance. Figures 5(a) and 5(b) show the magnetoresistance measured at $T$ = 40 K and its first derivative, respectively, in which we can easily see a marked difference between the film grown on YAP and those grown on the others.

The film grown on YAP exhibits a clear steplike magnetoresistance like bulk crystals do [5]. At $T$ = 40K, $\Delta R(H)/R(0)$ shows a steep decrease that begins at $H$ = 2 T and almost ends at $H$ = 6 T. If we define the WF transition field as the field where the first derivative shows a minimum, we obtain $H_{WF}$ = 4 T for this film as is indicated by an arrow in Fig.5(b).

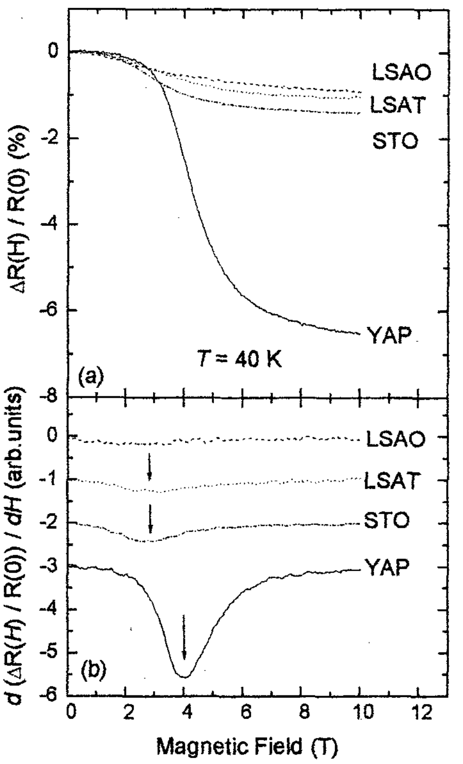


Fig.5 (a) Field dependence of the in-plane magnetoresistance of the films grown on different substrates. (b) The first derivative of the magnetoresistance shown in (a). Arrows indicate the position of the WF transition.

Such a steplike magnetoresistance behavior is strongly suppressed in the other films. At $T$ = 40 K and $H$ = 10 T, $\Delta R(H)/R(0)$ reaches only -1.1% (LSAO), -0.9% (LSAT), and -1.4% (STO), which are far smaller than that observed for the film grown on YAP. Nevertheless, one can find a trace of the WF transition in the films grown on LSAT and STO. Figure 5(b) shows that the WF transition still occurs at $H_{WF} \approx 2.5$ T in these two films, even though the critical field is lower than that for the film on YAP.

The weak ferromagnetism is more strongly suppressed in the film grown on LSAO, and the first derivative of the magnetoresistance becomes almost field-independent. We can no longer obtain clear $H_{WF}$ from Fig.5(b). If one carefully analyzes Fig.5(b), $H_{WF}$ is still discernible around $H$ = 1.5 T. However, this value is even lower than the $H_{WF}$'s of the films grown on LSAT and STO, and we conclude that the weak ferromagnetism is most strongly suppressed in the film grown on LSAO.

3. DISCUSSION

Whatever the origin of this peculiar magnetoresistance is [5,12], the WF transition is a clear evidence of the finite spin canting out of the $CuO_2$ plane that is inherent in the corrugated structure of $CuO_2$ planes. The present results indicate that the film grown on YAP actually has a corrugation in the $CuO_2$ plane similar to that in bulk crystals. The experimentally determined $H_{WF}$ (= 4 T) implies that the film grown on YAP is almost identical to bulk crystals, because single crystals show almost the same transition field [5]. However, it should be noted that the transition width across $H_{WF}$ is broader in this film than in reported crystals [4,5]. This suggests a spatial inhomogeneity of the transition field due to a finite lattice misfit between the crystal and the substrate.

The significant suppression of the spin canting by changing substrate symmetry from orthorhombic to tetragonal (or cubic) implies that the lattice symmetry of the substrates is a predominant factor to control the corrugation structure of the $CuO_2$ plane. However, the magnetoresistance of the films grown on LSAT and STO requires some explanations. The *c*-axis lengths of these films are shorter than that of the bulk crystals implying that the tensile strain is actually applied to the films, and thus we may expect the symmetry of $CuO_2$ planes to become tetragonal following the substrate symmetry. On the other hand, Figs.5(a) and 5(b) clearly show the presence of a weakened but finite step like magnetoresistance suggesting that the corrugation in the $CuO_2$ plane survives. One of the possible scenarios to explain these results is a twin structure: The films are separated into orthorhombic domains even though their orthorhombicity is not as large as in the films grown on YAP. We need further investigation on this problem.

In contrast to the cases of STO and LSAT, the weak ferromagnetism almost disappears in the film grown on LSAO. This is probably because that the lattice misfit of LSCO to LSAO is smaller than those to STO and LSAT [13]. We emphasize that not only the tensile strain but also the compressive strain is helpful in removing the corrugation from the $CuO_2$ plane. This result cannot be simply understood because the presence of corrugation implies that the $CuO_2$ plane has already been compressed by a rather small $La_2O_2$ blocking layer. With respect to this point, our results strongly indicate that the substrate symmetry is also essential for the removal of corrugation, and that the tetragonal substrate works well for these particular LSCO thin films.

4. SUMMARY

To summarize, we have found that the in-plane magnetoresistance of antiferromagnetic LSCO thin films is strongly dependent on the substrate material. The difference in the magnetoresistance behavior is attributed to the change in the corrugation structure of the $CuO_2$ plane. Within our experiments, the flattest

$CuO_2$ plane is obtained in films grown on $LaSrAlO_4$(001) substrates. It is suggested that not only the lattice parameters of the substrate but also its symmetry plays a significant role in removing the corrugations.

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[13] We calculate the misfit both along the *a* and *b* axis for LTO LSCO. By using the lattice parameters of bulk samples [10], the misfit is 0.85% (|| *a*) and 1.60% (|| *b*) to LSAO, -2.10% (|| *a*) and -1.37% (|| *b*) to LSAT, and -2.99% (|| *a*) and -2.26% (|| *b*) to STO. The smallest misfit is achieved to LSAO both along *a* and along *b*.

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