JOURNAL OF APPLIED PHYSICS VOLUME 95, NUMBER 11 1 JUNE 2004

Self-interference of charge carriers in ferromagnetic $SrRuO_3$

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(Presented on 8 January 2004)

We report a systematic study of the low-temperature electrical conductivity in a series of $SrRuO_3$ epitaxial thin films. At relatively high temperature the films display the conventional metallic behavior. However, a well-defined resistivity minimum appears at low temperature. This temperature dependence can be well described in a weak localization scenario: the resistivity minimum arising from the competition of electronic self-interference effects and the normal metallic character. By appropriate selection of the film growth conditions, we have been able to modify the mean-free path of itinerant carriers and thus to tune the relative strength of the quantum effects. We show that data can be quantitatively described by available theoretical models. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1682785]

Transport phenomena are very intriguing in strongly correlated systems. More specifically, metal transition oxides (MTO) show a plethora of phenomena such as high-T_C superconductivity and non-Fermi-liquid behavior in cuprates, or colossal magnetoresistance in manganites. A common feature in MTO compounds is the higher resistivity as compared to normal metals, which leads, applying the simple Drude approach, to a very short mean free path l that even under certain conditions approaches the Ioffe-Regel limit, i.e., $kl \approx O(1)$, where k is the momentum of the carriers.

The semiclassical Boltzmann picture is successfully applied in systems in the weak disorder limit, i.e., $kl \gg O(1)$, describing adequately the scattering of carriers by all type of perturbations that break the crystalline order such as impurities, interstitials, vacancies, phonons, magnons, and so on. However, a different approach, accounting for the full wavelike nature of the carriers, must be developed to understand the transport properties in systems at the strong disorder limit.[1–3] In the case of oxides, the mean free path can be close to the Fermi wavelength, so that $l \approx \lambda_F$, and weak localization effects (WLE) are expected to be relevant due to the self-interference of the carrier wave functions.

WLE can account for the presence of minima in the resistivity versus temperature curves at low temperature, frequently observed in different compounds and which cannot be explained using the semiclassical Boltzmann picture. On one hand, as the temperature decreases, the wave coherence of the carriers is enhanced, and WLE are reinforced, leading to an enhancement of the resistivity. On the other hand, as the temperature rises, the wave coherence is progressively lost and WLE are weakened, while the normal metallic behavior is recovered.

$SrRuO_3$ (SRO) is a ferromagnetic metal with a Curie temperature of $T_C \approx 160$ K, which is commonly used as electrode in technological devices.[4,5] In this article we report on the analysis of the low-temperature transport properties of epitaxial $SrRuO_3$ thin films grown on $SrTiO_3(001)$ (STO) substrates, assuming a WLE model.

In the past, WLE has been assumed as a model for the low-temperature transport properties in a variety of materials, including doped semiconductors, metal–metal oxide composites, alloys, and thin metallic films (see Ref. 1 for a review). More recently, some authors have studied ferromagnetic thin films, and there are reports on percolating Ni films,[6] on Fe films,[7] and on Co-doped FeSi alloys.[8] It must be emphasized that in most of these experiments, the mean free path has been modified by the introduction of impurities into the system and it is not obvious which is the precise impact on the electronic structure of the explored materials. On the other hand, in spite of WLE being suggested in $La_{0.8}Sr_{0.2}MnO_3$ single crystals, there is no conclusive work about the presence of WLE in oxides,[9] the main difficulty being the possible masking contribution of spin-dependent intergranular tunneling transport.

In this article we would like to overcome some of these difficulties by taking advantage of the sensitivity of strongly correlated systems and, in particular, of oxides to structural disorder. This property shall allow tuning their properties without introduction of foreign atoms into the structure. Next, intergranular spin dependent tunneling is expected to be less relevant in non-half-metallic ferromagnetic oxides. From both points of view, $SrRuO_3$ seems to be an ideal candidate to explore.

The samples were grown by pulsed laser deposition with a KrF excimer laser ($\lambda = 248$ nm, $\tau = 34$ ns). The oxygen pressure during the deposition was set to 0.1 mbar and the substrate temperature to 750 °C. Other experimental details can be found elsewhere.[10–12] The thickness of the nanometric films reported here was accurately determined after a calibration of the deposition rate by measuring thicker films by small angle x-ray reflectometry.

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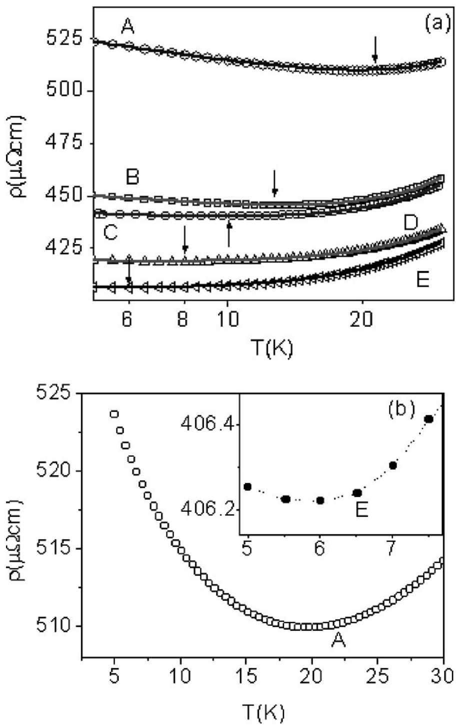

FIG. 1. (a) Plot of the temperature dependence of the resistivities of films varying the growth conditions. Curves *B* and *C* correspond to films with $t=6$ nm grown on nonvicinal and on vicinal (2°) substrates, respectively. Curves *A*, *D*, and *E* correspond to films cooled from the deposition temperature at different cooling rates (*A* and *D*, slow rate; *E*, fast rate). Arrows indicate the position of the minima. The fittings of the ρ-T curves to Eq. (3) are also shown. (b) Films *A* and *E* are plotted in a linear scale.

A common feature of the SRO films at this thickness is the presence of minima in the ρ-T curves, as can be seen in Fig. 1, corresponding to SRO thin films where the growth conditions were varied as explained below. Figure 1(a) is plotted in a logarithmic scale to allow seeing the experimental data set of points, indicating the quality of the measurements. The minima appear clearer when plotted in a linear scale, as in Fig. 1(b).

In order to gain an insight into the physical mechanisms lying behind the presence of such phenomena, we observe that as the residual resistivity ρ_0 increases, the temperature at which the minima appear is shifted to higher values [see Fig. 1(a)]. Put in another way, microstructural disorder enhances the physical processes that trigger the presence of such minima. Moreover, assuming a Drude model, we can estimate the mean free path from the low-temperature conductivity $\sigma=ne^2 l\lambda_F/h$, where n is the density of carriers, estimated as $n\approx 1\times 10^{22}\,\mathrm{cm}^{-2}$ from Hall effect measurements, and $\lambda_F\approx 4.5$ Å is the Fermi wavelength.[11] The estimated mean free path of the samples studied here is $l\approx 5-6$ Å,[11] hence close to the Fermi wavelength. Therefore one should expect that in these conditions WLE could play a significant role in the transport properties.

Under specific growth conditions, the microstructural disorder induced in the samples during the growth can be modified. The theory predicts that the corrections to the conductivity at low temperature due to the WLE are of order[1–3]

$$\sigma=\sigma_0\left[1+\left(\frac{\lambda_F}{l}\right)^{3/2}\left(\frac{k_BT}{E_F}\right)^{1/2}\right], \qquad (1)$$

so that by varying the microstructural disorder induced by the growth, i.e., by introducing slight changes of the mean free path, WLE should be modified, being enhanced as disorder increases. In the following we shall explain how we could modify the film growth, in order to alter the growth-induced disorder in SRO thin films.

In a previous report we showed the presence of a growth mode transition from an initial three-dimension (3D) growth to a final two-dimension (2D) growth in SRO thin films grown on nominally zero-miscut STO(001) substrates.[12] The initial 3D growth induces the formation of elongated islands along well defined directions following the substrate steps. As the thickness increases, the islands eventually coalesce and thereafter the growth proceeds by a 2D step mechanism. The initial nucleation depends on the step density and, therefore, on the miscut angle of the substrates. Thus, one expects that by using substrates with different miscut angles, the growth process should change, and hence the amount of structural disorder induced by the growth should be modified. We have grown two series of SRO thin films, one on nominally nonvicinal STO substrates, the other on STO substrates with miscut angle $\alpha=2°$. It is experimentally known that the vicinality of the $SrTiO_3$ substrates has a strong influence on the growth of $SrRuO_3$ thin films.[5,13] In agreement with these findings, our results indicate that structural disorder is smaller in films grown on the substrates with miscut angle $\alpha=2°$. In Fig. 1(a) the temperature dependence of the resistivity of two films with thickness $t=6$ nm grown on substrates with different vicinality is plotted (films *B* and *C*). We observed that both the residual resistivity ρ_0 and the temperature $T_{\min}$ at which the minimum appears are higher for the film grown on the nonvicinal substrate (film *B*).

On the other hand, we have observed that the cooling rate to room temperature from the deposition temperature, holding all the rest of the growth parameters unchanged, affects the magnetotransport properties of the films.[10] We observed that films cooled in low pressure (<20 mbar) after deposition—slow cooling rate—have higher resistivities and lower T_C than those cooled in a high-pressure atmosphere (1 bar O_2 or Ar). This fact can be rationalized by the results reported from the analysis of the temperature dependence of the bulk SRO crystallographic structure by synchrotron x-ray diffraction.[14] This study reveals that a series of polymorphic transitions occur within the temperature range $300\,\mathrm{K}\leqslant T\leqslant 950\,\mathrm{K}$. Due to the nature of these transitions, the rate at which the cooling is achieved can play a significant role. We suggest that at the regions where the coalescence of the initial elongated islands takes place a polymorphic phase can be stabilized by strain relief, different from that present in the regions inside the islands. Thus, the amount of disorder could be influenced by the cooling rate after the deposition. The temperature dependence of the resistivity of films with thickness $t=6-8$ nm grown in different conditions (*A*, *D*, slow cooling rate, $t=8$ nm; *E*, fast cooling rate, $t=6$ nm) are plotted in Fig. 1(a). We observe that the films cooled in a low-pressure atmosphere have higher resistivity and its $T_{\min}$

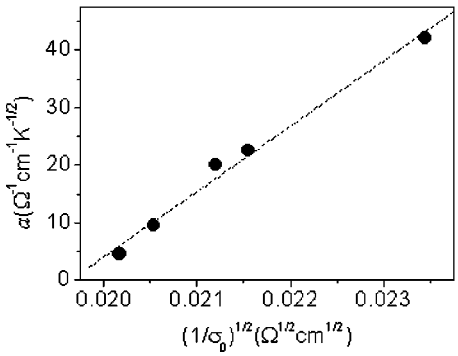

FIG. 2. The coefficient a of the weak localization $T^{1/2}$ term is plotted against $\sigma_0^{-1/2}$, where σ_0 is the residual conductivity. The data correspond to the samples reported in this article.

is higher. Apparently, the relevant parameter is the cooling rate, as no effect was observed by replacing O_2 by Ar when cooling in high-pressure atmosphere.

All the ρ-T curves appearing in Fig. 1(a) were analyzed using a WLE model including electron-electron interactions renormalized by self-interference effects. These effects can be described by[1–3]

$$\sigma = \sigma_0 + aT^{1/2}, \tag{2}$$

where the second term on the right accounts for interference effects. Since the analysis was performed in a temperature range $5\,\mathrm{K} \leqslant T \leqslant 30\,\mathrm{K}$ and assuming the validity of the Mathiessen rule, we have also added a T^2 contribution accounting for high-temperature scattering processes. Therefore, the analysis was carried using the expression

$$\rho = \frac{1}{\sigma_0 + aT^{1/2}} + bT^2. \tag{3}$$

From the analysis of the low-temperature resistivity, one can conclude that as the residual conductivity σ_0 decreases, the WLE coefficient a becomes more relevant, i.e., it is more significant as disorder increases (see Fig. 2). On the other hand, the theory predicts a relationship between a and σ_0,

$$a = \lambda_F^4 \left(\frac{ne^2}{h}\right)^{3/2} \frac{(2m^* k_B)^{1/2}}{h} \sigma_0^{-1/2}. \tag{4}$$

In Fig. 2 the coefficient a is plotted against $(\sigma_0)^{-1/2}$. From the slope of the fitting of the experimental results, one can get an estimate of the Fermi wavelength [see Eq. (4)], since the other parameters are known (the effective mass of the carriers can be estimated to $m^* \approx 3.7 m_e$ from Ref. 15). From this analysis it comes out that $\lambda_F \approx 2$ Å, which is close to the theoretic value $\lambda_F \approx 4.5$ Å, hence supporting the consistency of the analysis. However, further precise measurements under zero and applied magnetic fields are needed to have a thorough understanding of the low-temperature transport properties of our SRO thin films.

In summary, we have analyzed the low-temperature transport properties of epitaxial SRO thin films with thickness $t \approx 6-8$ nm. At these temperatures, minima were observed in the ρ-T curves and their presence was explained by weak localization effects. This analysis was justified by the fact that the mean free path of carriers is close to their Fermi wavelength. The results reported here emphasize the sensitivity of strongly correlated systems to disorder. As SRO is a common material used in technological devices, these results are of relevance for the understanding of the properties of multilayered heterostructures.

The authors acknowledge financial support from the MCyT (Spain) Project Nos. MAT2002-04551-C03 and MAT2003-4161.

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