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Hall-effect studies in $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattices

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We measured the resistivity and the Hall coefficient R_H in a series of $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ multilayers. We found no systematic change of the transport properties with decreasing layer thicknesses down to one unit cell. The resistivity and R_H evaluated for the $YBa_2Cu_3O_7$ layers are slightly higher than in bulk material, suggesting a small decrease of the carrier density in the multilayers. However, we observe no change in the Hall number $1/eR_H$ when the thickness of the $YBa_2Cu_3O_7$ layers decreases, so that the lowering of T_c observed in the superlattices cannot simply be related to a change in the carrier density. Furthermore, we find that the temperature dependence of R_H is very similar to that of bulk materials.

The superconducting oxides are anisotropic materials and their properties, both in the normal and in the superconducting state, reflect the anisotropy of their structure. It is generally accepted that the charge carriers are holes in the CuO_2 planes, and the Fermi surface of $YBa_2Cu_3O_7$, if there is one, is expected to have little dispersion along the *c* axis. Important questions here are to what extent the charge transport is a property of the individual CuO_2 planes and how important the coupling between the planes is for the superconducting properties. It has been shown recently[1-3] that it is possible to grow $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattices with layer thickness as thin as one unit cell. $PrBa_2Cu_3O_7$ is a semiconducting material, isostructural to $YBa_2Cu_3O_7$, so the $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ multilayers can be considered as a stack of thin conducting $YBa_2Cu_3O_7$ layers electrically disconnected by $PrBa_2Cu_3O_7$ sheets. These layered materials give us the opportunity to study the superconducting and normal-state properties of very thin $YBa_2Cu_3O_7$ layers. In particular it has been found[1-3] that the critical temperature T_c decreases as the thickness of the individual $YBa_2Cu_3O_7$ layers in the superlattices decreases, an effect whose origin is currently under investigation.

In this paper we report a study of the resistivity and the Hall-effect carried out on a series of $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ multilayers. The aim of our work is to determine whether the charge density of the CuO_2 layers is modified as the $YBa_2Cu_3O_7$ thickness decreases, and also to evaluate the effects of the interfaces on the normal-state properties. In fact a possible explanation of the above mentioned T_c decrease could be that the proximity of the semiconducting $PrBa_2Cu_3O_7$ induces a charge doping of $YBa_2Cu_3O_7$ across the interface.

The preparation of our superlattices is described in detail in Ref. 1. Briefly, we use a dc magnetron sputtering technique with $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ targets. Highly *c*-axis-oriented multilayers have been obtained both on MgO and $SrTiO_3$. X-ray analysis of the satellite peaks reveal that there is very little mixing between the $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ layers.[4] TEM analysis[5] shows clearly the layer modulation and very sharp interfaces between the layers. In the following we shall refer to the multilayers by the nominal thickness of the $YBa_2Cu_3O_7$ unit cell which we assume for simplicity to be a multiple of 12 Å (i.e., for example, 12/12 means one unit cell of $YBa_2Cu_3O_7$, one unit cell of $PrBa_2Cu_3O_7$, repeated several times). For this study we prepared a series of superlattices with equal thicknesses of the $YBa_2Cu_3O_7$ and the $PrBa_2Cu_3O_7$ layers.

For transport measurements we used the standard van der Pauw method with typical ac currents of 20 μA at 84.2 Hz. The Hall voltage was measured in magnetic fields, parallel to the *c* axis, up to 5 T and it was linear in magnetic field.

In Table I we report the critical temperatures T_c's of four multilayers. T_c is defined using the criterion 10% of the normal-state resistance. We find that T_c for the superlattices is lower than in $YBa_2Cu_3O_7$, but significantly higher than in the corresponding $Y_{0.5}Pr_{0.5}Ba_2Cu_3O_7$ alloy ($T_c = 21$ K from Ref. 1). This rules out the possibility of strong layer interdiffusion in our samples. Furthermore, if there would be some layer mixing due to a broadening of the interfaces, this would give rise to a systematic change of the transport properties with the decrease of the layer thicknesses. So, to further investigate this point, we studied the Hall effect for various modulation wavelengths Λ (Λ being twice the thickness of the individual $YBa_2Cu_3O_7$ layers for the series considered here). In Fig. 1 we show the Hall coefficient R_H measured on multilayers for temperatures above T_c. In this figure the data are evaluated disregarding the layer modulation, i.e., we used the whole film thickness to calculate R_H. This plot shows that all these R_H-vs-T curves almost overlap. In particular, there is no systematic change in the absolute value or in the temperature dependence of R_H due to the decrease of the layer thickness, at least within our experimental accuracy. The overlapping of the data is due to the fact that the ratio of the number of $YBa_2Cu_3O_7$ and $PrBa_2Cu_3O_7$ layers

TABLE I. Transport properties of multilayers, $YBa_2Cu_3O_7$ and $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy films. Note that, for the superlattices, the values of ρ, R_H, and n_H reported in this table are evaluated taking the thickness of the $YBa_2Cu_3O_7$ layers only.

Sample specification	ρ(300 K) ($\mu\Omega$ cm)	ρ(300 K)/ρ(100 K)	R_H(120 K) (10^{-9} m^3/C)	R_H(250 K) (10^{-9} m^3/C)	dn_H/dT (10^{19} $cm^{-3}T^{-1}$)	T_c (10%) (K)
$YBa_2Cu_3O_7$	260	2.4	0.98 ± 0.2	0.55 ± 0.10	3.67	84.5
96/96	305	1.8	1.37 ± 0.3	0.73 ± 0.15	3.11	74.0
36/36	290	1.3	1.42 ± 0.3	0.70 ± 0.15	3.53	67.5
24/24	285	1.7	1.48 ± 0.3	0.78 ± 0.16	2.97	68.0
12/12	395	1.2	1.50 ± 0.3	0.80 ± 0.16	2.82	50.3
$Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$	610	1.1	2.90 ± 0.5	1.64 ± 0.30	1.47	38.0

is the same (1:1) for all the samples. Based on this observation and on the structural studies, we analyze our data assuming our multilayers to behave as two independent resistances in parallel. As the resistivity of bulk $YBa_2Cu_3O_7$ is much lower than that one of bulk $PrBa_2Cu_3O_7$,[6] in the range of temperature considered here, we also assume that the whole current flows only in the $YBa_2Cu_3O_7$ layers. Consequently, the data that we report in the following are evaluated taking the thickness of the $YBa_2Cu_3O_7$ layers only, i.e., half of the whole film thickness. We anticipate that the transport properties of the layers are close to those of bulk $YBa_2Cu_3O_7$ and this supports the previous assumption. We also note that it is possible to show, by simple electrodynamic considerations, that the Hall coefficient of the superlattice is determined only by the $YBa_2Cu_3O_7$ layers as long as the conductivity of $PrBa_2Cu_3O_7$ is much lower than that of $YBa_2Cu_3O_7$.

In Table I we report the resistivity ρ measured at room temperature in our multilayers. Comparing these data with those obtained in one of our $YBa_2Cu_3O_7$ films and an $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy film we find that the multilayer resistivity at 300 K is slightly higher than the value measured on an $YBa_2Cu_3O_7$ film and much lower than that measured in the $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy. Furthermore, we do not observe any systematic change of ρ(300 K) related to the layer thickness, although the resistivity of the 12/12 superlattice is higher than the other ones. The latter result is probably due to the fact that the layer

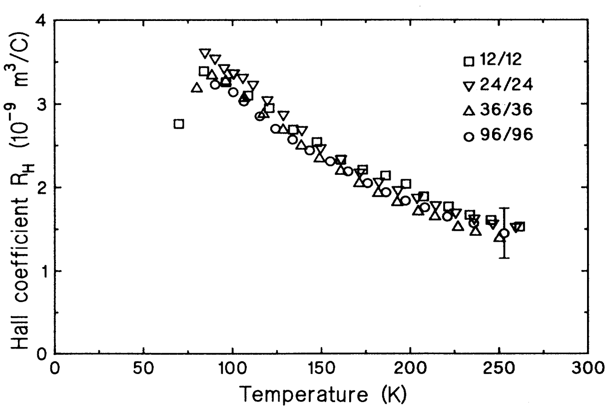

FIG. 1. Hall effect as a function of temperature for $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ (Å/Å) multilayers. Note that the absolute values of R_H are calculated taking the whole film thickness.

structure does not extend continuously along the whole specimen, but probably contains step discontinuities.[5] This kind of defect is crucial for the thinnest layers because they give rise to abrupt changes in the current paths. Another important feature of the multilayer resistivity is that the slope of the ρ-vs-T curves is always positive even for layer thickness as small as one unit cell. In Table I we report the resistance ratio ρ(300 K)/ρ(100 K) measured in our samples. We found ρ(300 K)/ρ(100 K) values between 1.2 and 1.8 for the multilayers with no evident systematic change related with the layer thickness. However, these values are lower than those found in $YBa_2Cu_3O_7$ films, i.e., the ρ-vs-T curves are flatter in the multilayers than in $YBa_2Cu_3O_7$. A flattening of the ρ-vs-T curves has been also observed in Nb/Cu and Nb/Al multilayers[7] and it was ascribed to the scattering with the interfaces. In our $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ superlattices the flattening of the ρ-vs-T curves occurs randomly and it seems mainly determined by material imperfections, as, for example, the aforementioned step discontinuities. However, the fact that the ρ-vs-T curves always have a positive slope indicates that the scattering with the interfaces is much less important than in multilayers made out of isotropic metals.

In Table I we also reported the Hall coefficient R_H measured at 120 and 250 K in our multilayers. A systematic study of the Hall effect in $Y_{1-x}Pr_xBa_2Cu_3O_7$ alloys has been reported by Matsuda *et al.*[6] They found that R_H increases when Y is progressively substituted by Pr and this suggests that Pr can give rise to a hole filling effect in $YBa_2Cu_3O_7$. However, it is not clear at the moment whether Pr acts as a simple electron donor in a rigid-band model, or if it induces a localization of the free carriers in the CuO_2 planes. In our multilayers we find that R_H is slightly higher than in $YBa_2Cu_3O_7$ and lower than the values measured in the $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy. The difference between the value of R_H found in multilayers and in $YBa_2Cu_3O_7$ could be due to some layers of low quality that do not contribute to the transport so the total layer thickness is smaller than the nominal value used to calculate R_H. However, this seems unlikely in our case, as we found almost the same increase of R_H in all the multilayers. Another possible origin of the increase of R_H could be that the carrier density is lower in the layers than in the bulk material. Note that a lower carrier density in the multilayers is also consistent with the resistivity data that we discussed previously. We have already shown in Fig. 1 that there is no systematic change of the Hall-effect related to the layer thickness and this seems to exclude a possible doping arising from substitution of Pr for Y. We rather propose that the R_H increase is related to the fact that the layers undergo a mechanical strain that is only weakly thickness dependent for such thin layers. This strain arises from the mismatch between the $YBa_2Cu_3O_7$ and the $PrBa_2Cu_3O_7$ lattices. For small modulation wavelengths the superlattices grow coherently and are strained.[8] For the multilayers studied here, relaxation of the strain by misfit dislocations are expected to occur for wavelengths larger than ~300 Å. This strain may have an influence on the oxygen ordering in the $YBa_2Cu_3O_7$ layers and this may lead to a slightly different carrier concentration than in the bulk material. It has been reported that T_c in $YBa_2Cu_3O_7$ scales with the Hall number $n_H = 1/eR_H$.[6,9] One may therefore think that the T_c variation in our multilayers might be related to a change of n_H. For instance, we note that the T_c of the 96/96 multilayer fits with the data reported in Ref. 9 using our experimental determination of n_H. However, the important result of this paper is that n_H is practically the same for all the superlattices investigated here, while their T_c ranges from 50 to 74 K. Although the relationship between n_H and the carrier density is not completely established, it appears reasonable to conclude, from the constancy of n_H, that the carrier concentration is the same in all our multilayers. Thus the T_c decrease from the 96/96 to the 12/12 multilayer cannot be understood as resulting only from a change in the carrier density and other mechanisms are necessary to explain this behavior.

Another interesting feature of the $YBa_2Cu_3O_7$/$PrBa_2Cu_3O_7$ multilayers is shown in Fig. 2 where we plot the temperature dependence of the Hall number n_H measured in our samples. We can compare these curves with those of the $YBa_2Cu_3O_7$ film and a $YBa_2Cu_3O_7$ crystal as well as the $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy film. These plots show that the linear temperature dependence of n_H, that is usually found in $YBa_2Cu_3O_7$, is not substantially changed by the decrease of the layer thickness in the superlattices. In Table I we report the slopes dn_H/dT of the n_H-vs-T curves obtained by linear fitting between 120 and 270 K. It results that dn_H/dT for the multilayers is slightly lower than for $YBa_2Cu_3O_7$ but higher than for $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy. No systematic change related to the layer thickness is observed. In Fig. 2 we can also see that the n_H-vs-T curves measured in multilayers point to small positive values at $T=0$ comparable to those found in our best $YBa_2Cu_3O_7$ films and crystals. These features of the n_H-vs-T curves measured in multilayers are consistent with the fact that there can be a slightly lower carrier density than in $YBa_2Cu_3O_7$.

The linear temperature dependence of the Hall number is not yet understood for the superconducting oxides and several models have been proposed. Clayhold *et al.*[10] suggested that it is related to the "unusual" ground state of the carriers and they noted that it could be a feature of materials with high T_c. Our results show that there is no correlation between the slope of the n_H-vs-T curves and T_c, as we have found almost the same linear behavior in all the multilayers regardless of their T_c. In the framework of the transport theory for fermionic system of carriers[11,12] any temperature dependence of the Hall number can arise both from the anisotropy of the carrier scattering and also from the temperature dependence of the $df/d\epsilon$ (f being the Fermi distribution function). If we ascribe the linear temperature dependence of n_H to anisotropic scattering of the carriers, we can conclude from our experiments that the scattering with the interfaces is not very important in the $YBa_2Cu_3O_7$/$PrBa_2Cu_3O_7$ superlattices as there is no systematic change of the n_H-vs-T

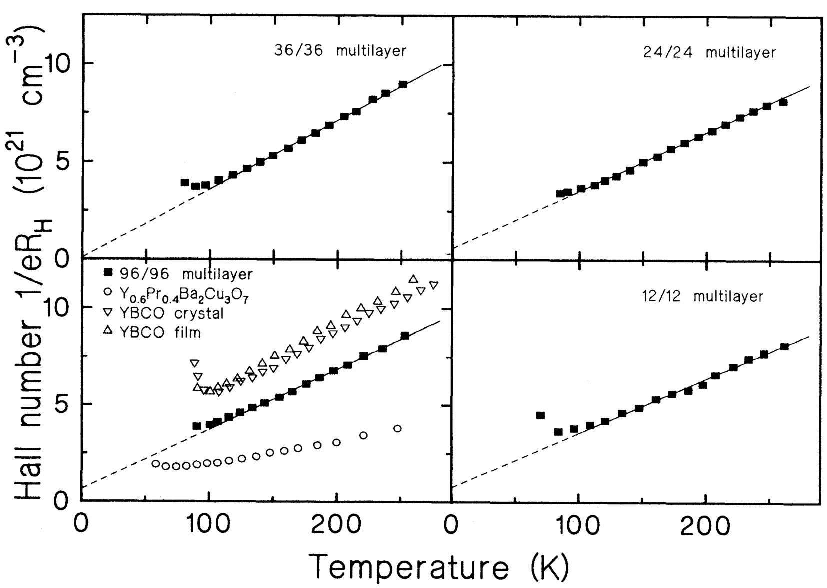

FIG. 2. Temperature dependence of the Hall number $1/eR_H$ for the multilayers, $YBa_2Cu_3O_7$ (film and crystal) and $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ alloy film. The crystal data are taken from Ref. 13. The straight lines represent the linear fitting of the data between 120 and 270 K. For the superlattices R_H was calculated using the thickness of the $YBa_2Cu_3O_7$ layers only.

curves due to the decrease of the layer thickness. This is also consistent with the resistity behavior of the multilayers that we discussed before. Finally, since the thinnest $YBa_2Cu_3O_7$ layers have a Hall-effect behavior very similar to that of $YBa_2Cu_3O_7$ crystal, we conclude that our results support the idea that the temperature dependence of n_H is essentially a feature of a quasi-two-dimensional system.

In conclusion, we have measured the normal-state resistivity and Hall effect in a series of $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ multilayers with layer thicknesses as small as one unit cell. The layer transport properties are very similar to the behavior of the bulk material and we do not observe any evident effect due to the presence of the interfaces. In particular, the Hall effect of the various multilayers is identical. This shows that the reduction of the thickness of the individual layers is not accompanied by a change in the carrier density.

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