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# Sign reversal of the Hall resistance in the mixed-state of  $La<sub>1.89</sub>Ce<sub>0.11</sub>CuO<sub>4</sub>$  and  $La<sub>1.89</sub>Ce<sub>0.11</sub>(Cu<sub>0.99</sub>Co<sub>0.01</sub>)O<sub>4</sub>$  thin films

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## ABSTRACT

The transport properties of  $La<sub>1.89</sub>Ce<sub>0.11</sub>CuO<sub>4</sub>(LCCO)$  and  $La<sub>1.89</sub>Ce<sub>0.11</sub>(Cu<sub>0.99</sub>Co<sub>0.01</sub>)O<sub>4</sub> (LCCO;Co) supercon$ ducting thin films are investigated. When the external field H is applied along the crystallographic c-axis, a double sign reversal of the Hall voltage in the mixed state of LCCO:Co thin films is observed whereas a single sign reversal is detected in LCCO. A double sign reversal of the Hall signal in LCCO can be recovered if the magnetic field is tilted away from the plane of the film. We find that the transition from one to two of the Hall sign reversal coincides with the change in the pinning from strong to weak. This temperature/ field induced transition is caused either by the magnetic impurities in LCCO:Co or by the coupling between the pancake vortices and the in-plane Josephson vortices in LCCO. These results are in agreement with early theoretical and numerical predictions.

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#### 1. Introduction

Hall effect is considered as a powerful method to probe the Fermi surface of metallic compounds in general, and particularly useful to identify the nature of the charge carriers in non-magnetic systems [\[1–5\]](#page--1-0). However, in the mixed state of superconductors, the Hall voltage is mainly determined by the vortex motion along the direction of the bias current flow [\[6\].](#page--1-0) In these systems, a long standing unsolved issue is the presence of a sign reversal in the Hall voltage when changing either temperature or magnetic field.

There have been many experiments and theoretical studies focused on this so called anomalous Hall effect. For example, sign reversals has been reported in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  (YBCO) crystals [\[7,8\]](#page--1-0) and La $_{2-x}$ Sr $_x$ CuO $_4$  thin films [\[9\]](#page--1-0), and double sign reversal and even triple sign reversal have been observed in  $Tl_2Ba_2CaCu_2O_8$  (TBCCO) [\[10\]](#page--1-0) and  $HgBa<sub>2</sub>CaCu<sub>2</sub>O<sub>6</sub>$  thin films [\[11\]](#page--1-0), respectively. Several models, based on two-band [\[12\]](#page--1-0), thermal fluctuation [\[13\]](#page--1-0), pinning effect [\[6,14\],](#page--1-0) and vortex interaction [\[15,16\],](#page--1-0) have been proposed to interpret the Hall anomalies. In electron-doped high- $T_c$  superconductors, a distinct feature revealed by the angular-resolution photoemission experiments [\[17\]](#page--1-0) is the coexistence of the holeand electron-bands near the optimal doping region, which may account for the sign reversal of the Hall resistivity for temperatures above the superconducting transition  $T_c$  [\[18\]](#page--1-0). However, for explaining the presence of sign reversals below  $T_c$ , it is still necessary to discuss them in terms of vortex pinning mechanisms which have been demonstrated to play a key role [\[14,15\]](#page--1-0).

Indeed, in a seminal work, Wang and Ting [\[19\]](#page--1-0) demonstrated that the Hall resistivity  $\rho_{xy}$  as a function of the magnetic field in the flux flow regime exhibits a sign change due to backflow currents arising from pinning forces. However, Vinokur et al. [\[6\]](#page--1-0) on the basis of a phenomenological model which included pinning and thermal fluctuation, concluded that the magnitude of the Hall angle  $\Theta_H$ , does not depend on the pinning. Whether pinning is necessary for the anomalous Hall effect or not is still a fundamental question, although there is clear experimental evidence indicating that the abnormal Hall effect is indeed pinning dependent [\[7\]](#page--1-0).

Based on their former work, Wang, Dong, and Ting (WDT) developed a unified theory for the flux motion [\[14\]](#page--1-0) by taking into account vortex pinning and thermal fluctuations which could explain both the single and the double sign reversals of the Hall resistivity due to different pinning strength. The WDT prediction has been further confirmed by Zhu et al. by a numerical simulation [\[20\].](#page--1-0) Their work has clearly shown that the double sign change of  $\rho_{xy}$  versus the temperature appears in the weak pinning environment, while the single sign reversal occurs in the strong pinning system. Importantly, the authors have also found that there are two distinctive weak pinning configurations which can achieve the double sign reversal of  $\rho_{xy}(T)$ , i.e., one possesses low density of the pinning centers but with strong individual pinning potentials, whereas the other consists of a high pinning concentration of weak individual pinning potentials. An elegant experimental study by Kang et al. [\[7\]](#page--1-0) unambiguously showed the relevance of the pinning strength for the sign reversal of the Hall resistance





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by irradiating a sample with heavy-ion to progressively increase the pinning strength. What it is still lacking is a more tunable pinning which allows one to go from the strong limit to the weak limit. In this case, one may expect a transition from the single sign reversal to the double sign reversal in the Hall resistivity  $\rho_{\rm xv}$ .

In the present work, we present experimental evidence on the relevance of the pinning strength in determining the number of sign reversals in the Hall voltage. We show that despite the fact that LCCO thin films exhibit only one sign reversal with the applied field perpendicular to the ab-plane, a second sign reversal emerges either when rotating the magnetic field and maintaining the same perpendicular component or by changing the pinning strength by substituting Co for Cu in order to obtain  $La<sub>1.89</sub>Ce<sub>0.11</sub>(Cu<sub>0.99</sub>Co<sub>0.01</sub>)O<sub>4</sub>$ (LCCO:Co). These two different methods show that the transition of the Hall sign reversals from single to double, coincides with the change in the pinning environment from strong to weak. Our experimental results are in agreement with previous prediction based on analytical and numerical investigations [\[14,20\]](#page--1-0).

## 2. Experimental details

All the LCCO thin films used in the present work were fabricated by a dc magnetron sputtering method [\[21,22\]](#page--1-0). The transport measurements of both LCCO and LCCO:Co thin films were carried out by using a commercial Quantum Design PPMS-14. More details about the film preparation and the measurement setup can be found elsewhere [\[18,21–26\]](#page--1-0). All the thin films were patterned by photolithography and subsequent ion milling, into bridges of  $2100 \mu m$  long, 100  $\mu m$  wide and with six terminals. The critical transition temperatures  $T_{c0}$  for LCCO and LCCO:Co are  $\sim$ 24 and 13 K as reported in our previous work [\[23\]](#page--1-0), respectively. The upper critical magnetic field  $H<sub>c2</sub>(0)$  for LCCO is about 10 T [\[18\].](#page--1-0)

### 3. Results and discussions

Let us start by demonstrating experimentally that the tilting of the magnetic field with respect to the plane of a thin LCCO film leads to significant changes in the pinning properties as well as the sign reversal of the Hall resistivity  $\rho_{xy}$ . In order to investigate this effect we study the Hall resistivity  $\rho_{xy}$  and longitudinal resistivity  $\rho_{xx}$  as a function of the perpendicular field  $B_z$  by either varying the strength but maintaining the field orientation perpendicular to the *ab*-plane (mode A) or by changing the angle  $\theta$  between the field and the ab-plane with constant field intensity (mode B). During the measurements, the applied current  $j$  in the  $ab$ -plane is always perpendicular to the magnetic field, i.e.,  $j\bot B$ . This method allows us to discern the effects arising purely from the out of the plane component of the applied field from those originated by the in-plane field.

First we would like to point out that in the LCCO sample and for  $T > T_c$ , both  $\rho_{xy}(B_z)$  and  $\rho_{xx}(B_z)$  curves overlap with each other no matter whether the perpendicular component of the magnetic field changes its magnitude or its orientation [\[18\]](#page--1-0). This indicates that the contribution of parallel-field component  $(B_v)$  to the magnetoresistance is negligible in comparison with  $B<sub>z</sub>$ . The question naturally arises whether this is also true for the case of  $T < T_{c0}$ . In Fig. 1, isothermal curves of  $\rho_{xy}$  versus  $B_z = B|\sin\theta|$  for LCCO are plotted at 15 K (<T<sub>c0</sub>). Here, we find that  $\rho_{xy}(B_z)$  curve does not follow the scaling with  $B_z$  as well as the  $\rho_{xx}(B_z)$  curve as reported in Ref. [\[18\]](#page--1-0). Notice that  $\rho_{xy}(B_z)$  with **B** $\perp$ *ab*-plane has the maximum value at  $\sim$  3.5 T and drops to zero when the magnetic field is decreased down to 1.4 T without reversing its sign, whereas the  $\rho_{xy}(B)$  measurement via rotation of the field exhibits a clear sign change from positive to negative in the low field region.



Fig. 1. Hall resistivity  $\rho_{xy}$  versus  $B_z$  ( $B$ |sin $\theta$ |) in LCCO thin films at 15 K by different field changing modes. Inset: illustration of tilted vortices with JV representing inplane Josephson vortices and PV representing the pancake vortices.

This result indicates the importance of the in-plane component of the field in the mixed state. When the magnetic field is titled away from the c-axis, two different kinds of vortices appear in the samples [\[27–29\]](#page--1-0), namely pancake vortices with the field component perpendicular to the ab-plane, and Josephson vortices with the component parallel to the ab-plane, as shown in the inset of Fig. 1. By rotating the field from in-plane to out-of-plane, the vortices change from Josephson vortex to pancake vortex type and vice versa.

In principle, the pinning effect for the in-plane vortices is always stronger than that for out-of-plane vortices, i.e., it is harder to drive the Josephson vortices out of the ab-plane. Therefore, a hysteresis in the depinning field may appear [\[30\]](#page--1-0). Based on this consideration it is clear that changing the field orientation from  $\theta$  =  $-$  90° to  $\theta$  = 90°, we should be able to observe the difference between  $\rho_{xx+\theta}$  and  $\rho_{xx,-\theta}$  due to the different evolution processes of the vortices. Here,  $\rho_{xx,+\theta}$  and  $\rho_{xx,-\theta}$  correspond to the resistivity values with  $\theta$  varying from 0 $\degree$  to 90 $\degree$  and from  $-90\degree$  to 0 $\degree$ , respectively. The experimental data seem to support this idea. In [Fig. 2](#page--1-0)a, the difference between  $\rho_{xx,+\theta}$  and  $\rho_{xx,-\theta}$ , i.e.,  $\Delta \rho_{xx} = \rho_{xx,+\theta} - \rho_{xx,-\theta}$ , is plotted as a function of  $B|\sin\theta|$  at 15 K with fixed magnetic field strength  $H = 10$  T. The maximum  $|\Delta \rho_{xx}|$  can reach up to 0.03 m $\Omega$  cm at  $B|\sin \theta| \sim 1.8$  T  $(\theta \sim 10^{\circ})$ , which is about 18% of the normal-state resistivity. With the increase of the field component along the c-axis, the sample is gradually driven into the normal state and  $\Delta \rho_{xx}$  approaches zero. This demonstrates that the in-plane field component plays an important role in the mixed state.

In [Fig. 2](#page--1-0)b, the curves of  $\rho_{xx}$  versus B|sin  $\theta$ | are shown for different modes, i.e., either rotating the orientation or changing the strength of the field. It is clear that for the same  $B<sub>z</sub>$ , the critical vortex depinning field for mode A with  $H \perp ab$  is higher than that for mode B by rotating field, where there is additional field component parallel to the ab-plane. The most important feature here is that the critical vortex depinning field for  $\rho_{xx,+0}$  from 0° to 90° is obviously larger than that for  $\rho_{xx,-\theta}$  from  $-90^{\circ}$  to 0°.

It is important to mention that the  $\rho_{xx}$  curve obtained via mode A for  $H \perp ab$  overlaps with the one corresponding to  $-H \perp ab$ , which suggests that the  $B_z$  component solely should not result in the hysteresis. Meanwhile, if we only apply the in-plane field, Hall signal is not expected. So the interaction between Josephson vortices and pancake vortices is essential for the sign reversal of  $\rho_{xy}$  in the mixed state via rotation of the field. We would like to point out that the field corresponding to the maximum  $|\Delta\rho_{xx}|$  is just the field where the vortices move from the plastic to the elastic mode [\[31\]](#page--1-0) as seen from the  $d\rho_{xx}$  /dB(B) curve in the inset of [Fig. 2b](#page--1-0).

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