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The van der Pauw method of measurements in high-T_c superconductors

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Details on the transport measurements of high-T_c superconductors have been presented using the van der Pauw technique. Basic procedures to obtain good and reliable results in polycrystalline samples were discussed. The influence of heating rates and the direction of the applied magnetic field on the results of measurements has been examined. An unexpected nonzero transverse voltage at zero magnetic field was observed in the vicinity of the superconducting temperature transition (T_c). Measurements in two different magnetic field directions allowed one to calculate the symmetric and the asymmetric components of the Hall resistance. Those components were calculated for two different superconductor systems and analyzed in the framework of the recent theory about the longitudinal and transverse voltages in high-T_c superconductors.

1. Introduction

The measurement of electrical resistivity and the Hall effect in high-T_c superconductors (HTSC) remains a subject of great interest [1–5]. In particular, the unusual behaviour of the Hall effect in many high-temperature superconductors and in some conventional superconductors has become a persistent problem in understanding the flux motion in superconductors. To measure resistivity and the Hall effect employing standard methods, the samples need to be processed into a bar structure with two end contacts, for passing electrical current, and two pairs of thin protruding leads on each side of the bar, where the longitudinal and Hall voltages can be measured. In an ideal case, the Hall contacts should be correctly placed to avoid longitudinal contributions to the Hall voltage. Since the superconductor samples are usually very small and have no regular geometry, the fabrication of such contacts is more difficult.

In a normal state, the Hall effect in superconductors is antisymmetric in an external magnetic field, and should be zero at zero magnetic field. However, for real sam-

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ples we usually find a finite voltage at zero field which can superimpose on the Hall signal when a magnetic field is applied. This undesired contribution can result from either a geometrical mismatch in voltage contacts, a misalignment of the magnetic field and current directions, or the inhomogeneity of the sample. Several methods are used to eliminate or reduce that undesired contribution. One of them is the technique elaborated by van der Pauw (vdP) [6, 7]. The van der Pauw technique enables the measurement of resistivities on thin samples with arbitrary shapes. In contrast to Hall-bar measurements, the vdP technique requires only four sufficiently small contacts placed at the circumference of an arbitrarily shaped (but simply connected) sample [6, 7]. The methods of reversing magnetic field and without reversing its direction may be used.

The method of reversing magnetic field is based on the fact that the Hall potential changes its sign (but not the absolute value) after reversing the magnetic field while the longitudinal potential remains the same. The non-Hall potentials can be thus eliminated after two measurements in both field directions (H, $-H$). Here, we have to keep in mind that the polarity of the injected current should be kept the same. So, the longitudinal and Hall voltages can be calculated by:

$$V_{XY} = \left(\frac{V_{AC,BD}(H) - V_{AC,BD}(-H)}{2} \right) \tag{1}$$

$$V_{XX} = \left(\frac{V_{AC,BD}(H) + V_{AC,BD}(-H)}{2} \right) \tag{2}$$

where the $V_{AC,BD}$ is defined as the voltage between points A and C when an applied current flows through contacts B and D (see Fig. 1).

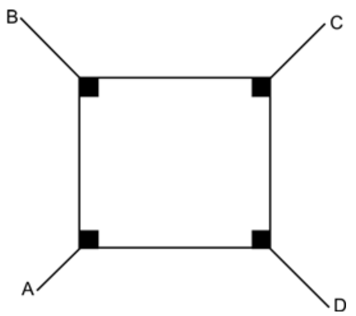

Fig. 1. Schematic view of the electrical contacts in the sample

In the method without reversing the applied magnetic field, the voltage due to the transversal electric field can be detected by permutation of voltage and current contacts (see Fig. 2). Any pair of contacts can be used as current contacts and the remaining pair as voltage contacts. From the reciprocity theorem it follows that $V_{AC,BD}(-H) = V_{BD,AC}(H)$ [8, 9]. Thus, the Hall and longitudinal voltages can now be reformulated as:

$$V_{XY} = \left(\frac{V_{AC,BD}(H) - V_{BD,AC}(-H)}{2} \right) \tag{3}$$

$$V_{XX} = \left(\frac{V_{AC,BD}(H) + V_{CD,AB}(-H)}{2} \right) \tag{4}$$

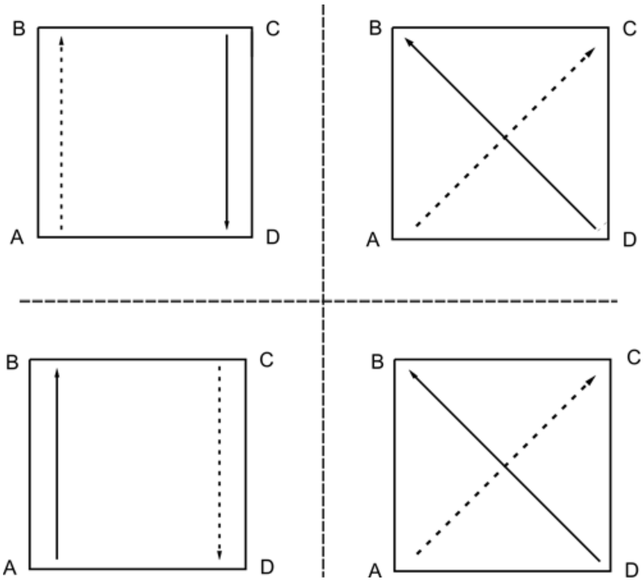

Fig. 2. Scheme of a four probe configuration used for the longitudinal (left) and Hall voltages (right) measurements. The solid and dashed arrows represent current and potential pairs of the contacts, respectively. In the top, the two configurations denote $V_{AB,CD}$ and $V_{AC,BD}$ and the other two below $V_{AB,CD}$ and $V_{BD,AC}$ voltages, respectively

Based on the methods described above, this work reports an important dc technique which uses the van der Pauw method for measurements HTSC polycrystalline sample. Firstly, a modified method has been presented. Basic procedures to obtain good and reliable results have been discussed. The influence of heating rates and of the direction of the applied magnetic field is described. The results are analyzed in the framework of the recent theory of the longitudinal and transverse voltages in high-T_c superconductors.

2. Experimental

Polycrystalline samples of $YBa_2Cu_3O_{7-\delta}$ were prepared by a solid state reaction. Samples of the $Bi_2Sr_2Ca_{1-X}Pr_XCu_2O_{8+\delta}$ superconducting system were also prepared with x = 0.2 and 0.3. X-ray powder diffractometry showed that all the samples are single phase. For more details about sample preparation and characterization see the references [10, 11].

In order to obtain a good voltage signal, the sample was mechanically polished until very thin square prism had been obtained. The contacts were placed in the corners as shown schematically in the Fig. 1. The electrical terminals were prepared using low resistance sputtered Au contacts (~ 0.1 Ω). Magneto-transport properties were studied using a Maglab Oxford system of 9T. The magnetic field was applied perpendicular to the sample surface. A constant current was applied using a programmable current source Keithley model 220, and the corresponding voltage was measured using a nanovoltmeter Keithley model 181. V_{XX} and V_{XY} were measured by means of the van der Pauw technique, with permutation of the voltage and current contacts. The contacts were appropriately switched using a computer-controlled relay circuit (Fig. 3). In this method, the Hall and longitudinal voltages were measured simultaneously at each temperature and each applied magnetic field. Data acquisition and calculation of V_{XX} and V_{XY} were done in real time using the LabVIEW software [12].

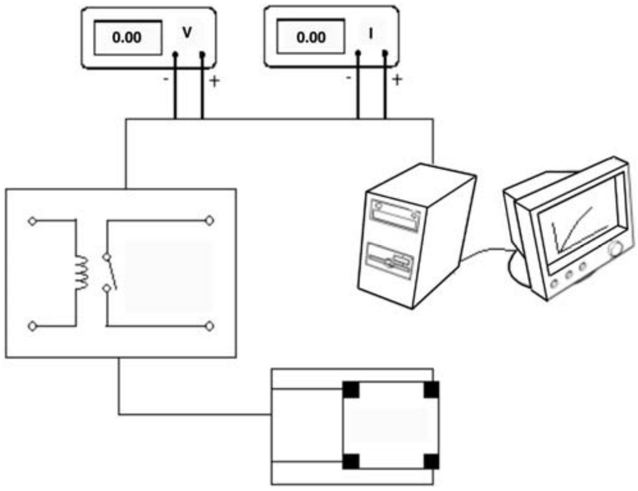

Fig. 3. Measurement setup for the modified switching contacts exploiting the van der Pauw technique. The contacts are switched using four different relays

Noise (thermopower effects, etc.) was eliminated by reversing the transport current polarity (I) in the samples under measurement. Thus, to obtain longitudinal (V_{XX}) and transverse (V_{XY}) voltages, eight voltage signals were measured. The V_{XX} and V_{XY} components were calculated based on the combination of eight records, which can be expressed as

$$V_{XX} = \frac{\left(V_{AB,CD}(I) - V_{AB,CD}(-I)\right)\left(V_{BC,AD}(I) - V_{BC,AD}(-I)\right)}{4} \quad (5)$$

and

$$V_{XY} = \frac{\left(V_{AC,BD}(I) - V_{AC,BD}(-I)\right)\left(V_{BD,AC}(I) - V_{BD,AC}(-I)\right)}{4} \tag{6}$$

By the method without field reversal, all voltages may be detected due to the transversal electric field. However, using Eq. (6), we can only partially compensate for those voltages added to the Hall effect. In this case we should also revert the magnetic field to separate the asymmetric and symmetric part of the Hall effect.

3. Results and discussion

Separating the Hall effect signal from its resistive offset signal is hard enough under normal circumstances, but it becomes even more challenging when the resistivity is drifting rapidly, as happens in materials transforming from metallic to superconductor behaviour. An important prerequisite for the validity of Eqs. (5) and (6) is that the measurements of the eight components are made under the same conditions, especially at the same temperature. Thus, the van der Pauw measurement of high-T_c superconductors requires consideration of some additional factors such as the influence of heating conditions of the sample during the measurement.

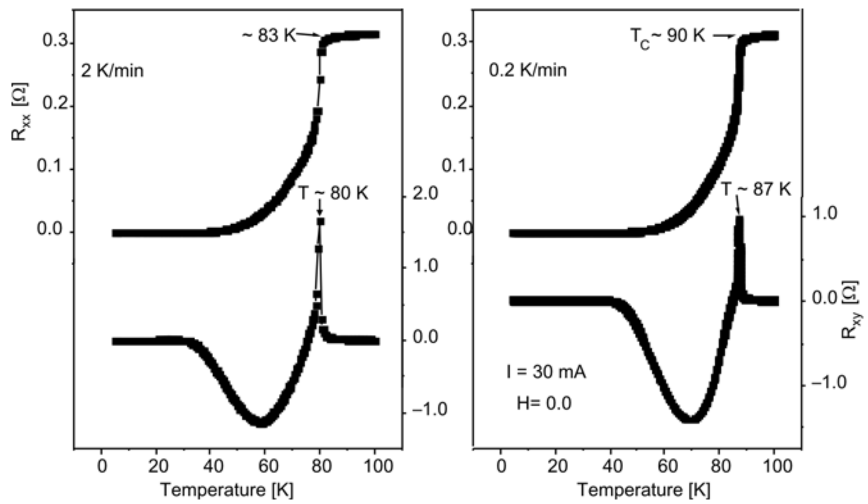

Fig. 4. Longitudinal resistance (upper curves) and transverse resistance of $YBa_2Cu_3O_{7-\delta}$ (lower curves) in function of the temperature measured for the various heating rates at $B = 0$, and $I = 30$ mA

We have performed a number of tests focused on the temperature stabilization of the sample. Figure 4 presents longitudinal (R_{XX}) and transverse (R_{XY}) resistances in function of temperature for the sample of $YBa_2Cu_3O_{7-\delta}$ measured at various heating rates under zero applied magnetic field. Measurements were taken during heating at the rates of 0.2 K/min and 2 K/min. At 2.0 K/min, the onset of the superconducting transition is far from the well-reported critical temperature for $YBa_2Cu_3O_{7-\delta}$ ($T_{ci} \approx 90$ K). However, at 0.2 K/min, T_{ci} is ca. 89 K which suggests that the measurement was performed under thermal equilibrium. Note that R_{XX} is essentially independent of the heating rate but the longitudinal resistance moves towards lower temperatures for higher heating rates. However, in the R_{XY} signal, both the intensity and the temperature dependence are altered with the change in the heating rate. The peak intensity (positive) at T_c increases while the second peak intensity (negative), at lower temperatures, decreases as the heating rate increases. Also both peaks move towards lower temperatures at 2 K/min. Based on these results, we conclude that lower heating rates are crucial for reliable transport measurements in superconductor materials, especially for Hall effect measurements.

The fact of occurrence of the Hall resistance at zero magnetic field is worth noting. In the mixed state, the Hall resistance in superconductors results from hydrodynamics vortices produced by the current flowing through the sample in an external magnetic field [1–5]. Then, in zero magnetic field we should not observe the appearance of a transverse voltage. However, transverse voltages in zero magnetic field have been observed in various HTSC materials [1, 5, 13–17]. Near T_c, a free vortex in the mixed state was observed which can be generated without the application of an external magnetic field. In this case, vortex–antivortex pairs may be excited by thermal fluctuations in the superconducting state, or induced by transport current passing through the sample on the opposite sides of the sample [13–17]. From Fig. 4, we can clearly see nonzero longitudinal transverse voltage close to T_c at zero magnetic field. The V_{XY} curves are non-zero close to the critical temperature with the maximum value approximately in the middle of the longitudinal superconducting transition, which has been reported in the literature [14, 15]. Moreover, a sign change from positive to negative is observed as the temperature decreases. In a true superconducting state ($R = 0$ in the longitudinal resistance) and in the normal state ($T > T_c$), no finite transversal voltage was detected. These results can be explained based on induced vortex–antivortex pairs applying the guiding vortex model [15]. The theory of the guiding vortex was proposed to explain the even effect in superconductors which assumes the existence of a new force acting on the vortex. This force (the guiding force) impels the vortex to move only in a given direction that is determined by the direction of the pinning potential valley. However, the nature of this pinning potential has not been completely solved. One of the promising models is the intrinsic pinning model, which assumes that the origin of the guiding forces is due to the layered structure of the HTSC [18]. Other mechanisms such as grain boundary guiding in polycrystalline materials for example, should be kept in mind. This model seems to offer a consistent explanation of our results.

In general, transverse voltage consists of three components: a component coming from the geometrical misalignment of the contacts, a Hall voltage component, and a component originating from the guided motion of vortex and antivortex. The geometrical misalignment can be detected by normal state measurements in zero magnetic field. In our case, in the normal state, no transverse voltage was detected at zero applied magnetic field. Therefore, the transverse voltage in zero magnetic field cannot originate from a geometrical misalignment of the contacts. The other two components can be distinguished by comparing the voltages measured in two opposite magnetic field directions: while in the Hall voltage, the sign of the measured voltage depends on the applied magnetic field direction because the vortex motion is only governed by the Lorentz force: in the case of the guided vortex–antivortex motion, it is independent of the field direction. This difference gives us the chance to distinguish between generation mechanisms of these two voltages. Figure 5a shows the transverse voltage in function of the temperature measured in the same sample that was presented in Fig. 4 under magnetic field in positive (H = 9 T) and negative (H = –9 T) directions.

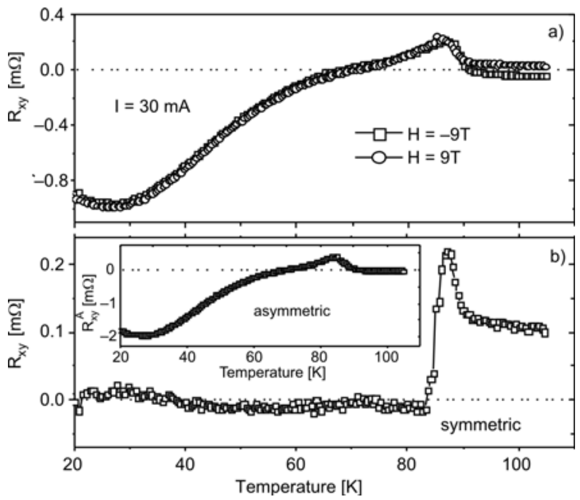

Fig. 5. Transverse resistance in function of the temperature measured in the sample of $YBa_2Cu_3O_{7-\delta}$ at the applied magnetic field in positive (H = 9 T) and negative (H = –9 T) directions (a); and symmetric and asymmetric transverse resistances calculated from the data of Fig. 4a (b)

From Figure 5a we can observe two distinct behaviours: above T_c (ca. 90 K), R_{XY} is dependent on the direction of the applied magnetic field; below T_c, the transverse resistance is independent of its direction. Using R_{XY} measurements in both directions of the magnetic field, we can calculate the symmetric (R_{XY}^S) and asymmetric (R_{XY}^A) resistance components. To obtain the symmetric transverse resistance which is due to the applied magnetic field, we should calculate $R_{XY}^S = (R_{XY}(H) - R_{XY}(-H))/2$, where $R_{XY}(H)$ and $R_{XY}(-H)$ are measured under magnetic field applied in positive and negative directions, respectively. The asymmetric transverse resistance was calculated as $R_{XY}^A = (R_{XY}(H) + R_{XY}(-H))/2$. The results for both components are presented in Fig. 5b. In the normal state ($T > T_c$), the symmetric component presents a positive signal, as observed for several superconductor materials. In the superconducting state ($T < T_c$) no transverse voltage was detected for this value of magnetic field. Thus, we can say that this component presents the true Hall resistance effect. The inset (Fig. 5b) shows the asymmetric component which can be due to guided motion of vortex and anti-vortex, similar to Hall measurements in zero applied magnetic field.

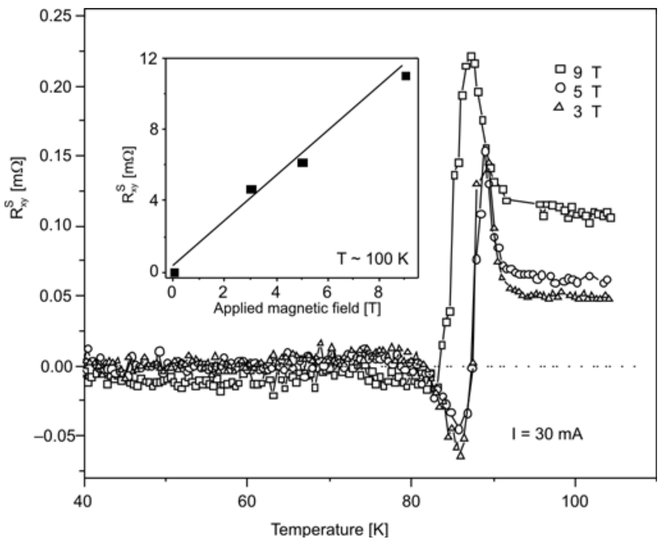

Fig. 6. R^S_{XY} in function of temperature for the same sample $YBa_2Cu_3O_{7-\delta}$ for various magnetic fields. The inset shows the transverse resistance in function of magnetic field at ca. 100 K

In Figure 6, we show other results for R^S_{XY} in function of temperature for the sample $YBa_2Cu_3O_{7+\delta}$ for various magnetic fields. In the normal state (see inset), the transverse voltage is positive and increases linearly as the strength of the applied magnetic field increases, as is expected of hole-doped high-T_c superconductors [19, 20]. The resistance changes from positive to negative below T_c for low and moderate magnetic fields. Higher fields suppress this sign reversal and restore $R_{XY} > 0$ near T_c, while at low temperatures R_{XY} tends to zero. There are several models to explain this effect, but common agreement has not been achieved up to now [3, 5]. In addition to the sign reversal near T_c, the R^S_{XY} measured for the sample $Bi_2Sr_2Ca_{0.8}Pr_{0.2}Cu_3O_{8+\delta}$ suggest the occurrence of a second sign reversal which has been observed in most high-T_c superconductors (see inset of Fig. 7) [21, 22]. The error is about 5%. Figure 7 shows longitudinal resistance (upper) and symmetric transverse resistance (lower), calculated from the results of the van der Pauw measurements at 5 T. In the R_{XX} we can see a broad superconducting transition which can be related to granular superconductivity. The R^S_{XY} is positive in the normal state and changes to negative around T_c. These sign reversals in the transverse component are believed to be related to the quasiparticle or vortex-core contributions which are associated with the normal state excitation, superconducting dissipation resulting from vortex hydrodynamics and superconducting fluctuations [22].

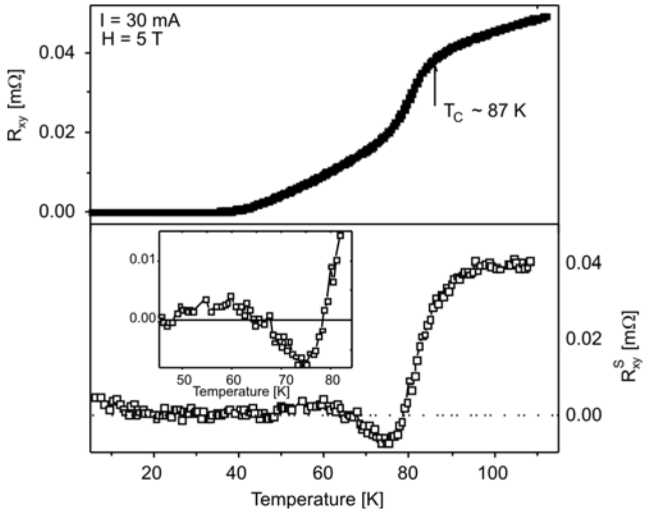

Fig. 7. Longitudinal resistance (upper) and symmetric transverse (lower) resistances calculated from the results of the van der Pauw measurements at 5 T, for the sample of $Bi_2Sr_2Ca_{0.8}Pr_{0.2}Cu_3O_{8+\delta}$. In the inset, the results suggest a double sign reversal of the R^S_{XY} in the vicinity of T_c

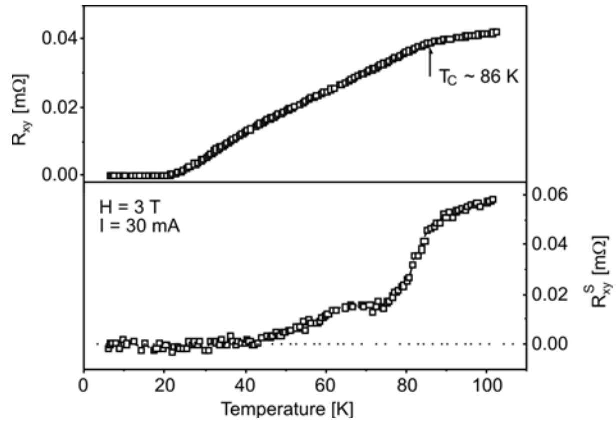

Fig. 8. Longitudinal resistance (upper) and symmetric transverse (lower) resistances calculated from results of the van der Pauw measurements at 3 T, for the sample of $Bi_2Sr_2Ca_{0.7}Pr_{0.3}Cu_3O_{8+\delta}$

Finally, the longitudinal resistance and symmetric transverse resistance, calculated from results of the van der Pauw measurements at 3 T for $Bi_2Sr_2Ca_{0.7}Pr_{0.3}Cu_3O_{8+\delta}$ are shown in Fig. 8. We have noted a very wide superconducting transition in the temperature dependence of R_{XX}. In contrast with that observed for $Bi_2Sr_2Ca_{0.8}Pr_{0.2}Cu_3O_{8+\delta}$

(see Fig. 7), no sign inversion was observed in the transverse component. This difference in behaviour probably reflects the difference in the flux dynamics and the nature of the pinning mechanism between these two superconductor materials. Similar behaviour is observed in some of the conventional high-T_c superconductors in the mixed state, but is not fully understood yet [23].

4. Conclusion

A modified van der Pauw technique has been presented to measure transport properties of HTSC superconductors. It was observed that a low heating rate is crucial for obtaining reliable transport measurements in high-T_c superconducting materials. It is necessary to carry out measurements in two opposite magnetic field directions in order to separate the transverse resistance components. We believe that this technique makes real-time Hall and longitudinal measurements practical in undergoing superconducting transitions.

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