Electrostatic Tuning of the Hole Density in $NdBa_2Cu_3O_{7-\delta}$ Films and its Effect on the Hall Response

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We have used the ferroelectric field effect in heterostructures based on superconducting $NdBa_2Cu_3O_{7-\delta}$ and ferroelectric $Pb(Zr_{0.2}Ti_{0.8})O_3$ to electrostatically modulate in a reversible and nonvolatile fashion the hole carrier density of the superconducting layer. Reversing the ferroelectric polarization induces a constant relative change in the resistivity and Hall constant of 9% and 6%, respectively, at all temperatures above the superconducting transition. The cotangent of the Hall angle displays a T^2 dependence with a slope that increases as the carrier density is reduced.

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The origin of the temperature-doping phase diagram of the high T_c superconductors, the relationship between the pseudogap and superconductivity, and the properties of low carrier density systems in general have been the subject of intense experimental and theoretical investigations [1,2]. In this context, the normal state properties of oxide superconductors are important and can contribute to the understanding of these complex materials. Soon after the discovery of cuprate superconductors, the anomalous normal state transport properties, which include a dc conductivity proportional to $1/T$ and a Hall angle proportional to $1/T^2$ [3–5], have been recognized as distinctive signatures of these materials. The identification of the exact role of the carrier concentration in the temperature dependencies of the longitudinal and transverse conductivities of these materials, however, is a difficult experimental task. Chemical doping, which is the most commonly used technique to change the carrier concentration, invariably introduces disorder, and often magnetic scattering, rendering the interpretation of the data difficult. In this Letter, we report on an experimental approach based on modulation of the carrier concentration using the electrostatic field effect. This approach allows us to identify the exact role of the carrier concentration on the Hall constant and the Hall angle temperature dependencies without introducing any structural disorder. We find that changing the carrier concentration electrostatically induces a constant relative change in the resistivity and inverse Hall constant as a function of temperature, pointing to a resistivity and Hall constant that are both inversely proportional to a temperature independent carrier density.

Field effect experiments are in principle straightforward. Applying an electric field to a metallic film creates a charge depletion or accumulation layer, whose characteristic thickness is given by the Thomas-Fermi screening length λ_{TF}. λ_{TF} depends only on the carrier concentration, and in oxide superconductors is typically on the order of a unit cell (1 nm) [6]. Substantial effects will thus be observed only in ultrathin films. "Classical" field effect experiments using a conventional dielectric have been used previously to shift the transition temperature of high T_c superconductors [7,8], to study vortex dynamics and critical currents [9], and, most recently, to induce superconductivity in C_{60} materials and organic compounds [10].

In a series of papers [11,12], we have demonstrated the feasibility of using the polarization field of a ferroelectric in epitaxial ferroelectric/superconductor heterostructures to modulate in a nonvolatile, reversible fashion the carrier density. Because the ferroelectric polarization of perovskite materials such as $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT) is large (up to 40 $\mu C/cm^2$) and can in principle deplete completely one unit cell of typical oxide superconductors, substantial modification of the carrier concentration can be achieved. We have used this approach to demonstrate reversible switching behavior between insulating and superconducting behavior in underdoped high T_c superconductors [12].

Figure 1 shows schematic top and side views of the devices studied for this work. Epitaxial $NdBa_2Cu_3O_{7-\delta}$-(NBCO)-PZT heterostructures were grown by rf magnetron sputtering onto (100) $SrTiO_3$ substrates. Typical thicknesses are 80–100 Å for NBCO and 1000 Å for PZT. X-ray finite size oscillations around the (00l) reflections of NBCO and PZT single layers indicate high crystalline quality and allow for the direct determination of the film thickness. Atomic force and scanning tunneling topography studies of the PZT and NBCO thin films demonstrate that these films display smooth surfaces. The root mean square roughness is 2–3 Å (over 5 μm × 5 μm) for 1000 Å PZT films [13] and ± one unit cell for 100 Å thick NBCO films (over 0.5 μm × 0.5 μm).

As depicted in Fig. 1, these bilayers have been lithographically patterned, allowing for the simultaneous measurement of the resistivity and Hall effect. For transverse resistivity measurements, the magnetic field parallel to the c axis is swept from -5 to $+5$ T while keeping the temperature constant to within 5 mK. To switch the ferroelectric polarization, we use the metallic tip of an atomic

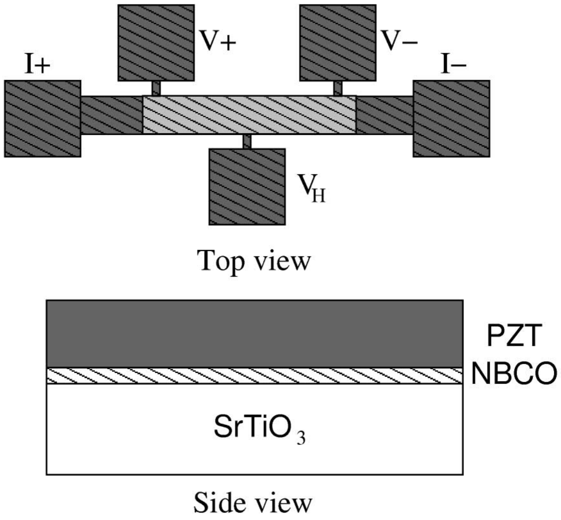

FIG. 1. Side view: Schematic view of the device; on a (100) $SrTiO_3$ substrate, a thin (001) NBCO layer and (001) PZT layer have been deposited. Top view: The path used to measure the resistivity and Hall effect. The dimensions of the patterned resistivity paths are 20 μm (width) by 80 μm (length). The light gray area is where the polarization is switched.

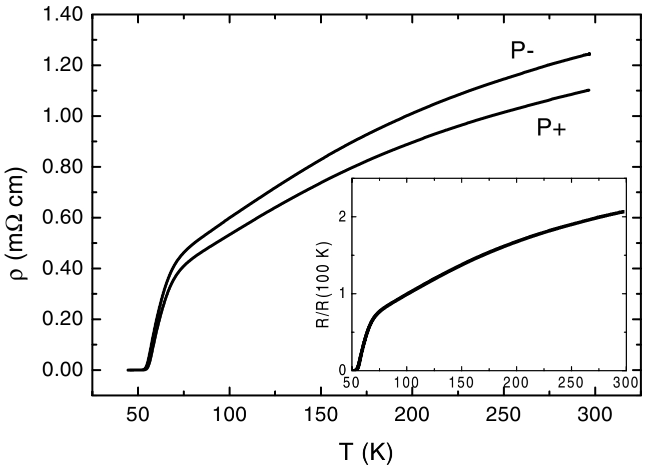

FIG. 2. Resistivity versus temperature for the two polarization states. P^+ is the polarization direction that adds holes to the superconducting layer. The change in resistivity is about 9% at room temperature. The shift in T_c is about 1 K. Inset: Normalized resistivities as a function of temperature.

force microscope (AFM) as a mobile electrode that is scanned over the whole area of the conducting path between the voltage contacts [11]. While scanning the tip, a constant voltage of $V_t = +10$ or -10 V is applied between the tip and the superconducting film, a procedure that has been used before to homogeneously pole large areas [13]. During the transport measurements, no voltage is applied across the ferroelectric, precluding spurious effects related to leakage currents or strain [14].

Figure 2 shows the influence of the ferroelectric polarization on the resistivity of the samples. The curve labeled P^+ corresponds to the polarization state that adds holes to the superconductor. For this direction of the polarization, the resistivity is lower, in agreement with the hole doped character of $R\text{Ba}_2\text{Cu}_3\text{O}_7$ (R = rare earth) compounds. The curve P^- is obtained after having reversed the polarization. We note that the ferroelectric polarization of PZT is temperature independent below room temperature [15]. The measured relative change in resistivity associated with the switching from P^+ to P^-, $\Delta\rho/\rho$, is about 9% at room temperature and decreases slightly to 8% at T_c. In the region of the superconducting transition, the change in T_c is about 1 K. The inset of Fig. 2 shows the two resistivity curves normalized at 100 K. We notice that the data rescale, as would be expected if the polarization is temperature independent and the resistivity is simply proportional to $1/n$, as in a free electron model.

Figures 3 and 4 show Hall effect measurements and are the central results of this paper. Figure 3a shows the inverse Hall constant as a function of temperature for the two polarization states. As is generally observed, R_H^{-1} depends

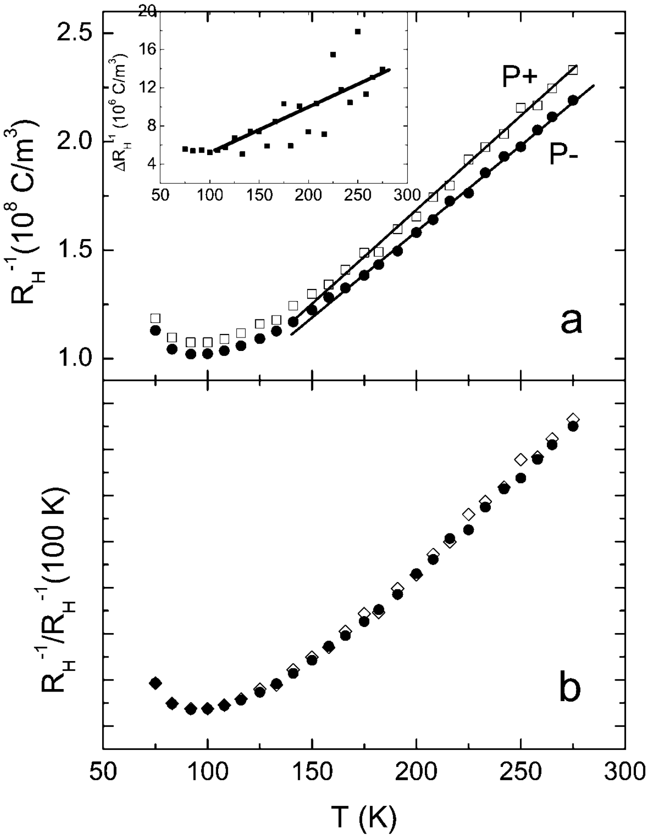

FIG. 3. (a) Inverse Hall constant R_H^{-1} as a function of temperature for the two polarization states. The solid lines are fits to the data in the linear region. Inset: Difference in the inverse Hall constant between the two polarization states as a function of temperature. The solid line is the difference between the two fits shown in (a). (b) Normalized inverse Hall constant versus temperature.

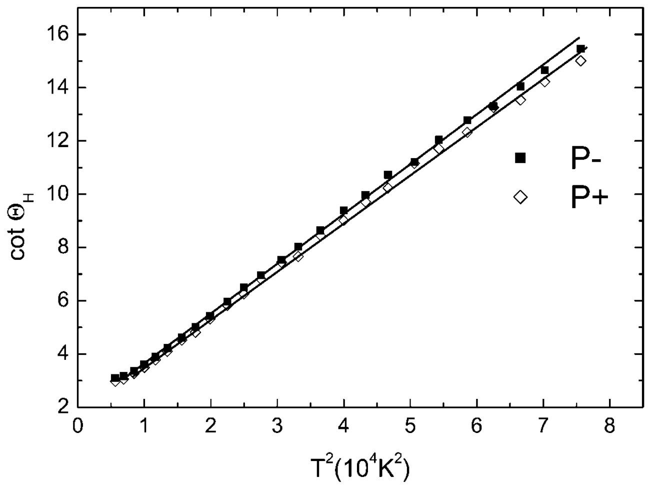

FIG. 4. $\cot\Theta_H$ as a function of T^2 for the two polarization states. The lines are a guide to the eye.

approximately linearly on temperature ($R_H^{-1} = AT + B$) over a large temperature region. In RBa$_2$Cu$_3$O$_7$ compounds, the linear region is generally observed down to T_c for optimally doped samples and down to the pseudogap temperature T^* for underdoped samples [16–18]. For the sample shown here, the value of R_H^{-1} and the critical temperature are typical of underdoped samples, and we estimate T^* to be around 150 K (the temperature at which R_H^{-1} deviates from a linear temperature dependence; see fit on Fig. 3a). As can be seen from the data, the ferroelectric field effect changes the Hall constant. The relative change in R_H^{-1} associated with the switching from P^+ to P^- is about 6% at room temperature and decreases to 5% at 100 K. In the simplest theory of the Hall effect, the inverse Hall constant is related to the carrier density n: $R_H^{-1} = qn$, where q is the charge of the carriers. In high temperature superconductors, the value of R_H^{-1} at a given temperature can be related to the doping level of the material (and its carrier concentration). The change in R_H^{-1} can thus be understood as due to the change in carrier concentration produced by the polarization reversal. The change in Hall constant also agrees with the sign of the carriers. The polarization state P^+ adds holes to the superconducting layer, decreasing the resistivity and increasing T_c. A rough estimate of the field effect at 100 K with $P \approx 10$ μC/cm^2 and $d = 10$ nm, gives $\Delta R_H^{-1} \sim 2 \times 10^7$ C/m^3, a value that agrees well with the observed change [19]. The estimate made here is based on average changes in the Hall constant (and resistivity) of the film, although the field induced change in carrier concentration is inhomogeneous. Since accounting for the inhomogeneous carrier distribution requires a detailed modeling of the Hall response (which would imply a detailed knowledge of the parameters entering the model), we will in the rest of the paper analyze the measured average changes in the resistivity and Hall constant upon polarization reversal.

Since the ferroelectric polarization of PZT is temperature independent below room temperature, one expects $\Delta R_H^{-1}(T)$ to be constant, since the induced carrier density change $\Delta n = \Delta P/qd$, where d is the film thickness, is constant. The experimental data in Fig. 3a show, however, that ΔR_H^{-1} is not constant as a function of temperature. In fact, it changes by a factor of 2 between room temperature and T_c, as does R_H^{-1} itself. This change is illustrated in the inset of Fig. 3a, where we have plotted $\Delta R_H^{-1}(T)$. As with the resistivity, the Hall data can be rescaled, as shown in Fig. 3b. We note that the rescaling is not affected by the opening of the pseudogap.

Figure 4 shows the cotangent of the Hall angle, $\cot\Theta_H$ ($\cot\Theta_H = \rho_{xx}/\rho_{xy} = \rho_{xx}/HR_H$) , as a function of temperature for the two polarization states. As observed by others for RBa$_2$Cu$_3$O$_7$ compounds [20], $\cot\Theta_H$ displays a T^2 temperature dependence down to 100 K, a result consistent with recent measurements on thin underdoped GdBa$_2$Cu$_3$O$_{7-\delta}$ films [18]. The T^2 temperature dependence of $\cot\Theta_H$ has been originally discussed in the theory of Anderson based on spin-charge separation [21]. Upon polarization reversal, since the relative increase in resistivity (about 9%) does not equal the decrease in inverse Hall constant (about 6%), the cotangent of the Hall angle (essentially the product of these two quantities) should be affected by the polarization change. Although small, this effect can be observed in the data, in particular at high temperatures. In chemical doping experiments, the slope of $\cot\Theta_H$ versus T^2 generally decreases with decreasing doping levels [17,22]. In our experiments, we observe that decreasing the carrier density increases the slope of $\cot\Theta_H$. This difference could be linked to the absence of induced structural changes or disorder in our electrostatic modulation of n.

We now discuss some of the implications of these results. While several models have been developed to explain the normal state transport properties of high temperature superconductors, recent photoemission spectroscopy measurements of the temperature evolution of the Fermi surface place restrictions on the possible theoretical scenarios [23]. In particular, the partial destruction of the Fermi surface for $T < T^*$ and the corresponding lack of a continuous Fermi contour in momentum space should be taken into account in the calculations of the transport properties. Stojkovic and Pines [24] have proposed a phenomenological model, where the dependence of the transport properties on the carrier concentration is made explicit. In their scenario, which is based on electronic spin interactions, the quasiparticles are more strongly scattered in areas of the Fermi surface intercepting the magnetic Brillouin zone boundary, which are termed “hot” spots, than in other parts, which are called “cold” regions (the hot spots are the areas of the Fermi surface at which the pseudogap and superconducting gaps open [25]). This anisotropic interaction gives rise to two scattering times, τ_{hot} and τ_{cold}, that have different temperature dependencies. The following expressions are found for the resistivity and Hall constant [26]:

$$\rho = \frac{1}{ne^2}\sqrt{\frac{m_{\mathrm{hot}} m_{\mathrm{cold}}}{\tau_{\mathrm{hot}}\tau_{\mathrm{cold}}}} \qquad R_H = \frac{1}{2ne}\sqrt{\frac{\tau_{\mathrm{cold}}/m_{\mathrm{cold}}}{\tau_{\mathrm{hot}}/m_{\mathrm{hot}}}}. \tag{1}$$

In this phenomenological model, there is no detailed physical description of the pseudogap, which is only taken into account through changes in τ_{hot}, leaving n unchanged. In our experiment, the changes observed upon reversing the polarization in ρ and R_H^{-1} agree with the estimated change for the carrier density n, and the rescaling properties of ρ and R_H are naturally explained by the theoretical expressions above, where the resistivity and Hall constant are inversely proportional to a temperature independent mobile carrier concentration. The temperature dependencies of ρ and R_H^{-1} then arise from the temperature dependencies of the scattering times, and changing n simply multiplies R_H^{-1} and ρ by a constant factor [27]. In theoretical approaches using a temperature dependent n [28,29], the rescaling is less transparent.

The difference observed between the relative change in ρ and R_H^{-1} is small, but has been systematically observed in all the samples investigated. We note here that the model of Hirsh predicts a difference in the relative change in ρ and R_H^{-1} due to a change in effective mass [28]. In the theoretical framework of Stojkovic and Pines, $\cot\Theta_H$ (proportional to ρR_H^{-1}) is equal to $2m_{\mathrm{cold}}/eB\tau_{\mathrm{cold}}$. This formula depends only on τ_{cold} and m_{cold} (and not explicitly on the carrier density).

A possible explanation of the small difference observed between the cotangent of the Hall angle for the two polarization states, about 3% (and thus the difference in the relative change of ρ and R_H^{-1}), is that the effective mass m_{cold} is increased when the carrier concentration is lowered due to increased Coulomb interactions between the quasiparticles since the system is closer to a Mott insulator, and/or that the magnetic fluctuations are stronger, leading to a smaller τ_{cold} [30]. Both effects lead to an increase of $\cot\Theta_H$.

In conclusion, we have shown that the ferroelectric field effect can be used to modulate in a reversible and nonvolatile way the normal state transport properties of oxide superconducting films. We find that electrostatic modulation induces a relative, temperature independent change in the resistivity and inverse Hall constant. These data find a natural explanation within the framework of the theory proposed by Stojkovic and Pines, where both the resistivity and the Hall constant are inversely proportional to a temperature independent mobile carrier concentration.

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