Correlation between incoherent phase fluctuations and disorder in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ epitaxial films from Nernst effect measurements

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Measurements of Nernst effect, resistivity and Hall angle on epitaxial films of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$(Pr-YBCO, $0\leq\ x\ \leq 0.4$) are reported over a broad range of temperature and magnetic field. While the Hall and resistivity data suggest a broad pseudogap regime in accordance with earlier results, these first measurements of the Nernst effect on Pr-YBCO show a large signal above the superconducting transition temperature(T_c). This effect is attributed to vortex-like excitations in the phase incoherent condensate existing above T_c. A correlation between disorder and the width of the phase fluctuation regime has been established for the YBCO family of cuprates, which suggests a $T_c \approx$110K for disorder-free $YBa_2Cu_3O_{7-\delta}$.

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The anomalously large Nernst voltage well above the zero-field T_c in hole-doped cuprate superconductors is now a well established experimental observation with a dominant view that it is due to vortex-like excitations in the phase uncorrelated superfluid above T_c.[1,2] Such excitations nucleate in the presence of an external field due to a non-zero pairing amplitude of incoherent phase at $T>T_c$ and drift down the thermal gradient generating a transverse voltage. The appearance of the Nernst signal on approaching T_c from above, therefore, marks the onset of a phase uncorrelated pairing amplitude. The observation of an enhanced diamagnetism near the onset temperature T_ν of the anomalous Nernst signal in some hole-doped cuprates strongly supports the vortex-like excitations scenario.[2,3] The fact that the regime of this large Nernst effect overlaps with the temperature range where a pseudogap is seen in the electronic excitation spectrum, somehow also suggests that the anomalous Nernst effect may be related to the pseudogap phenomenon, although counterviews also exist on the prescription of vortex-like excitations and correlation between Nernst effect and pseudogap phenomenon.[4,5] Rullier-Albenque *et al.*[6] have established a correlation between the width of the phase fluctuation regime over which a large Nernst voltage is seen and disorder in the CuO_2 planes induced by electron irradiation. The disordered samples show a wider range of phase fluctuations. However, high T_c cuprates can also be subjected to out-of-plane disorder by changing the ionic radius of the rare earth and alkaline earth sites while keeping the hole concentration fixed. The disorder works as a weak scatterer and reduces T_c substantially.[7]

The cuprate $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$(Pr-YBCO) presents a very interesting system to study the role of out-of-plane disorder on the regime of incoherent phase fluctuations in $YBa_2Cu_3O_{7-\delta}$ cuprates because the ionic radius of Pr^{3+} is larger by a factor of about 1.134 compared to the ionic radius of Y^{3+}. In this paper, we present the first measurements of the normal state Nernst Effect in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ over a broad range of composition. These data have been augmented by measurements of Hall angle and in-plane resistivity over a wide range of field and temperature. We note that while the zero-field superconducting transition temperature T_c drops with increasing Pr in a quasi non-linear manner as reported earlier,[8] the fluctuation regime $\triangle T_{fl}$=(T_ν-T_c) widens. Most remarkably, an interesting correlation emerges between T_ν and T_c in the YBCO family of cuprates with in-plane and out-of-plane disorder.

The c-axis oriented epitaxial $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$($x$=0, 0.1, 0.2, 0.3, 0.4) films of thickness about 2500 Å were fabricated on (100) $SrTiO_3$ substrates by pulsed laser deposition using a KrF excimer laser(λ=248 nm)with a typical repetition rate and energy density of 5 Hz and 2 J/cm^2 respectively, which yields a growth rate of 1.6 Å/second. The deposition temperature and oxygen partial pressure during film deposition were set 800 ^0C and about 400 mTorr respectively.

The in-plane resistivity and Hall effect measurements were done on the films patterned to a standard Hall bar in a Quantum Design PPMS with a 14 T magnet. The Nernst measurements were performed using a one-heater-two-thermometer technique. The sample was attached on one end to a copper block with a mechanical clamp and the other end was left free. A small chip resistor heater is attached on the free end, and a temperature gradient is created by applying a constant current to the heater. Two tiny Lakeshore Cernox thermometers are attached on the two ends of the sample to monitor the temperature gradient continuously. The Nernst voltage is measured with a Keithley 2001 multimeter with a 1801 preamp while the field is slowly ramped at a rate of 0.3 T/min between -14 T and +14 T($H\perp ab$). The system temperature was well controlled to give stability of the temperature of $\pm$1 mK, which enables us to perform a high resolution Nernst voltage measurement(typically ~10 nV in our setup). The temperature gradient is around 0.5-2 K/cm depending on the temperature of measurement, and the sample temperature is taken as the average of hot and cold end temperatures. The Nernst signal is obtained by subtracting negative field data from positive field data to eliminate any possible thermopower contribution. The Nernst signal was defined as

$$e_y = \frac{E_y}{-\nabla T} \quad (1)$$

where E_y is the transverse electrical field across the sample and $-\nabla T$ is the temperature gradient along its length.

Fig. 1 shows the temperature dependence of in-plane resistivity ρ_{ab} for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ films with Pr concentration from 0 to 0.4. The zero field superconducting transition temperature T_c decreases from 90 K for the x=0 film to about 40 K for the underdoped x=0.4 film in agreement with published work on these cuprates.[9,10,11] The in-plane resistivity increases with Pr content, which suggests a decrease of carrier concentration or carrier mobility due to out-of-plane disorder caused by Pr. While the resistivity $\rho_{ab}(T)$ remains metallic in the normal state, a small upturn in $\rho_{ab}(T)$ on cooling is seen in the x=0.4 film just above T_c, which may be due to charge localizations found in many other cuprates. The resistivity has a linear temperature dependence for the fully oxygenated Pr-free film, while it deviates from linearity on increasing the Pr doping. The temperature below which this deviation from linearity sets in is about 190 K in the x=0.1 film and gradually exceeds 300 K for the film with the highest Pr concentration. This nonlinear ρ_{ab} below 300 K for the higher Pr-rich samples indicates a pseudogap regime above T_c. This deviation of ρ_{ab}(T) from linearity on cooling is not monotonic. To illustrate this point we show in the inset of Fig. 1 the temperature derivative $d\rho_{ab}(T)/dT$ of all the films. The $d\rho_{ab}(T)/dT$ vs T plot of the films with $x\geq0.2$ goes through a maximum and the peak temperature shifts towards higher values with the increasing Pr content. This observation is consistent with the resistivity data of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ single crystals.[8] We note that the peak temperature T_{cr} is much lower than the pseudogap temperature, as found from the deviation of the linear resistivity. Sandu *et al.*[8] have identified this critical temperature T_{cr} in the ρ_{ab}(T) data of their $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ single crystals as a signature of the onset of dissipation due to thermally excited vortex loops. We will shortly compare T_{cr} with the onset temperature T_ν of the anomalous Nernst voltage, which is a direct indicator of vortex loop excitations. At this juncture, it is also worthwhile to point out that the overall behavior of the $\rho(T)$ of these films is similar to that observed by Convington and Greene[12] in their $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ films. The important consequence of Pr doping is a significant enhancement in resistivity without affecting the linear temperature dependence of ρ at T≥120 K. This

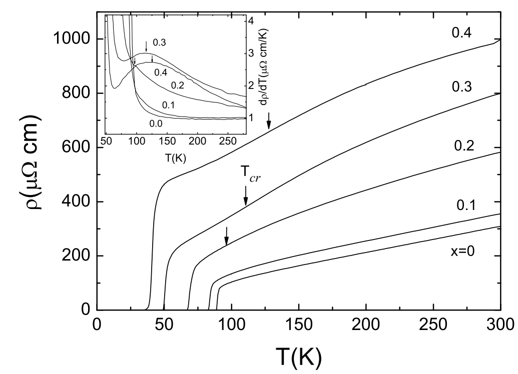

FIG. 1: In-plan resistivity as a function of temperature for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ films(x from 0 to 0.4). Inset shows the temperature derivative of the resistivity from the main panel for all the films. The arrows indicate the temperature where the peak of the derivative plot appears.

points towards enhanced scattering within CuO_2 planes without affecting the carrier concentration as seen in Zn doped YBCO. At larger Pr concentrations($x\geq0.5$), the $\rho(T)$ curves develop an S shape similar to that seen in oxygen deficient YBCO in the vicinity of superconducting transition. This is a signature of reduction in carrier concentration.

The temperature dependence of Hall resistivity for all the films was measured in a 14 tesla field for both field orientations. Consistent with prior work(see Ref. 7 and 8), the normal state Hall coefficient R_H first increases as the temperature is lowered from 300 K and then drops near the superconducting transition. At a given temperature, the Hall number decreases with Pr doping. It is about 5 times smaller at T=300 K in the x=0.4 film than for the Pr-free sample suggesting a strong localization of mobile holes by the local field of the Pr ions. The temperature dependence of the Hall angle $cot\theta(T)$ for all the films was calculated. We find that the $cot\theta(T)$ data can be fitted to the form $cot\theta = a + bT^n$ with n close to 2. A similar temperature dependence of $cot\theta(T)$ in Pr-YBCO samples has been observed in previous reports.[9,10,11] A deviation from the power law dependence of $cot\theta(T)$ on temperature is seen near T_c in all films. It has been suggested that this deviation could be related to the opening of the pseudogap.[13] To find the temperature T_H where this deviation starts, we plot $(cot\theta - a)/bT^n$ as a function of temperature for the films(Fig. 2) with x=0.2 and 0.3. As seen in the figure, $(cot\theta - a)/bT^n$ is a constant of order unity when the temperature is much higher than T_c. It starts to increase sharply below a critical temperature T_H ~105 K for these two Pr content films. We actually find that T_H remains nearly independent of the Pr concentration. Since T_H which is close to T_{cr} found from the resistivity, is much lower than the pseudogap temperature, it has also been argued[14] that the T_H scale may

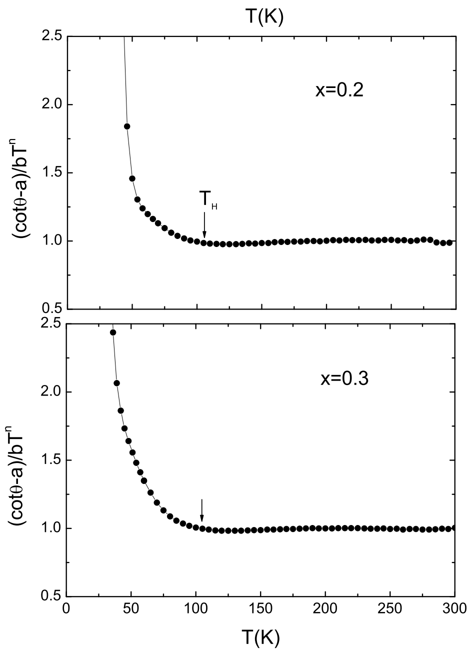

FIG. 2: $(cot\theta - a)/bT^n$(see text for details) versus temperature for x=0.2 and 0.3 films. The arrows mark the temperature T_H at which the deviation from the high temperature behavior starts.

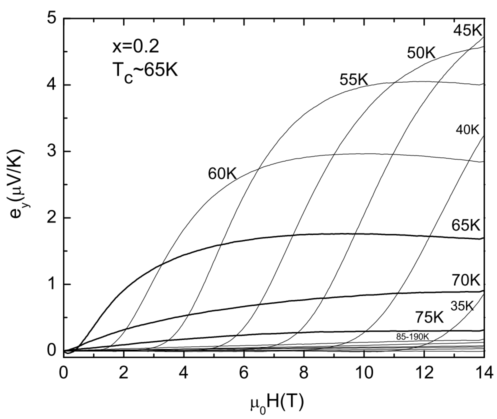

FIG. 3: The field dependence of the Nernst signal for the x=0.2 film at different temperatures.

be related to the onset of superconducting fluctuations or vortex-like excitations in the normal state.

The Nernst effect was measured in all Pr-substituted films. Fig. 3 shows the field dependence of the Nernst signal(e_y) at different temperatures for x=0.2 film. A qualitatively similar field dependence of e_y was observed for the other concentrations of Pr. These data are not

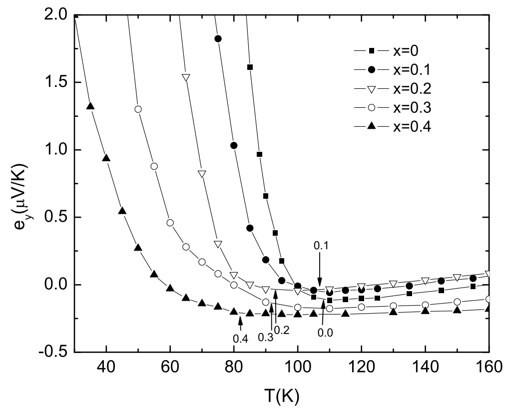

FIG. 4: The temperature dependence of the Nernst signal taken at 14 T for the films. The arrows show the temperatures at which the Nernst signal deviates from the high temperature background.

shown in Fig. 3 for the sake of clarity. The rapid rise of the Nernst signal for field less than the upper critical field (H_{c2}) observed for T<T_c is due to the motion of vortices driven by the temperature gradient. At higher temperatures(T>T_c), the Nernst signal remains sizable and has a non-linear field dependence. On increasing the temperature well beyond T_c, the signal e_y becomes extremely small. Here it tends to a negative linear field dependence, which typically is attributed to quasiparticles in the normal state.[2]

The temperature dependence of the Nernst signal taken at 14 T for all the films is shown in Fig. 4. As seen in the figure, this signal is extremely small for all the films in the high temperature range well above T_c. On decreasing the temperature, the Nernst signal starts to increase rapidly at a certain temperature T_ν, which depends on the Pr concentration. The temperature (T_ν) below which the Nernst signal rises rapidly above the high temperature normal state data is marked by arrows in Fig. 4.

The large Nernst signal observed in the temperature window of T_c and T_ν has been interpreted as evidence for vortex-like excitations or strong superconducting fluctuation in most of the hole-doped cuprates.[2] In Fig. 5, we show the characteristic temperatures T_{cr}, T_H and T_ν deduced from the measurements of resistivity, Hall angle and Nernst effect respectively along with the zero-field transition temperature T_c as a function of Pr concentration. We note that the onset temperature of the anomalous Nernst signal T_ν is lower than T_H and T_{cr}, and the difference between them increases in Pr-rich samples. It has been found[3] that in hole-doped cuprates, the onset temperature of the anomalous Nernst signal compares well with the temperature at which a fluctuating diamagnetism appears, it is clear that T_ν is a true indicator of the emergence of vortex like excitations in a phase in-

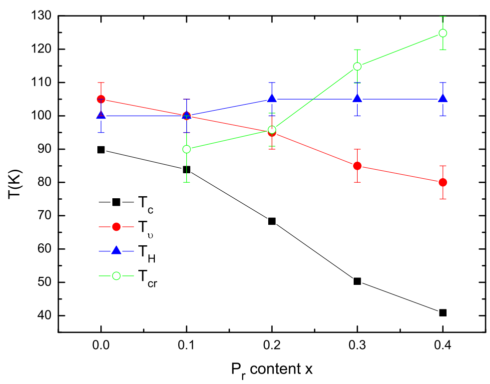

FIG. 5: (color online). Doping dependence of the temperature scales deduced from the temperature derivative of the in-plane resistivity(T_{cr}, open circle), Hall angle(T_H, solid triangle) and Nernst effect measurements(T_ν, filled circle), solid square is the superconducting transition T_c.

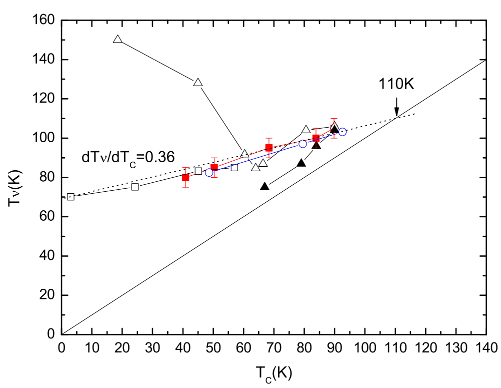

FIG. 6: (color online). Onset temperature T_ν of anomalous Nernst signal versus T_c for oxygen-doped and disordered YBCO. Solid square: Pr-YBCO, open triangle: oxygen-doped YBCO(ref. 16), open square: $YBa_2Cu_3O_{6.6}$ with electron irradiation(ref. 6), open circle: $YBa_2Cu_3O_7$ with electron irradiation(ref. 6) and solid triangle: Zn-YBCO(ref. 15).

coherent condensate. We note that while the T_c drops with increasing Pr, the width of the fluctuation regime $\triangle T_{fl}$(=T_ν-T_c) actually broadens.

The observed increase in $\triangle T_{fl}$ can be associated primarily with the out-of-plane disorder caused by the substitution of Pr at the Y sites of $YBa_2Cu_3O_7$. This site disorder may also have a spin component as the moment on the Pr sites can lead to pair breaking effects. Rullier-Albenque *et al.*[6] have studied the effects of in-plane disorder on the Nernst effect in $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_{6.6}$. They note that the fluctuation regime above T_c expands considerably with the disorder. In Fig. 6 we plot the T_ν vs T_c data of our films along with the results of Rullier-Albenque and coworkers. Quite remarkably, these data fall on a single curve with a slope $dT_\nu/dT_c \sim 0.36$. The figure also shows the characteristic temperature T_ν for Zn-doped YBCO.[15] It is known that zinc causes a strong in-plane disorder with a drastic suppression of T_c. The normalized temperature T_ν of the zinc doped YBCO also follows the general trend seen in Fig. 6. A simple extrapolation of the curve shown in Fig. 6 suggests that a disorder-free $YBa_2Cu_3O_7$ should have a T_c of 110K. In Fig. 6 we have also plotted the T_ν of the pristine oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ crystals.[16] Although these samples do not have any deliberately created in-plane or out-of-plane disorder there is some randomness in the occupancy of the plane site oxygen due to a non-zero δ. This positional disorder of oxygen should create local fluctuations in the potential seen by holes. Moreover, the hole concentration of these samples decreased with δ. The T_ν vs T_c curve for these samples shows a large deviation from the data for the disordered samples when the oxygen concentration falls below 6.6 per unit cell of YBCO. From these data it is clear that at least below optimal doping the phase fluctuation regime derives contributions from disorder as well as deficiency of mobile carriers. We expect that electron irradiation of $YBa_2Cu_3O_{7-\delta}$ with $\delta > 0.4$ would enhance their T_ν.

In summary, we have performed measurements of the Nernst effect, resistivity and Hall effect on Pr-substituted $YBa_2Cu_3O_{7-\delta}$ films. We find that an anomalous large Nernst signal survives above the superconducting transition temperature in the Pr-rich samples. This large Nernst voltage is attributed to vortex-like excitations in a phase incoherent superfluid existing above T_c. The regime of temperature over which these fluctuations prevail broadens with the Pr concentration. We attribute this effect primarily to the out-of-plane disorder caused by praseodymium, which is consistent with the measurements on other YBCO cuprates with in-plane and out-of-plane disorders. However, we do not completely rule out the contribution of reduced carrier concentration, particularly in the Pr-rich sample($x \approx 0.5$).

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