行政院國家科學委員會專題研究計畫成果報告
次攙雜高溫超導銅氧化物磁電傳輸性質與磁學性質之研究
An investigation of magneto-transport properties and magnetic properties of underdoped high $-T_c$ cuprates

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一、中文摘要

我們有系統地研究了鐠摻雜的本質性次摻雜124多晶樣品的磁傳輸性質與磁學性質。鐠的有效磁距隨著摻雜量增加而遞減，顯示了鐠4f與氧2p的軌道雜化效應隨著摻雜量的增加變得較為顯著。對於所有鐠摻雜的樣品，在磁化率倒數與溫度的關係圖中，在180 K均觀察到一個小轉折。這意味著此特徵溫度似乎不受軌道雜化效應的影響。另外，磁阻在低溫表現出異常的非線性的行為。這個有趣的現象可能與銅氧平面的磁學性質有關，亦可能是自旋能隙開啟的另一個表徵。

關鍵詞：本質性次摻雜，磁阻，軌道雜化，自旋能隙

Abstract

The temperatures dependences of magnetic susceptibility and magnetoresistance (MR) of intrinsically underdoped polycrystalline $Y_{1-x}Pr_xBa_2Cu_4O_8$ samples prepared at ambient oxygen pressure have been investigated. The effective moment of Pr obtained from χ(T) measurements varies from 2.81 μ_B to 2.46 μ_B as x changes from 0.1 to 1.0, indicating that hybridization between Pr *4f* and O $2p_\pi$ becomes more pronounced as Pr content increases. Another remarkable feature is a kink around 180 K in χ^{-1}(T) curve observed in all Pr-doped samples. It appears that this "characteristic" temperature is insensitive to the hybridized state which binds doped holes to Pr sites. Furthermore, MR measurements exhibit unusual nonlinear behavior, with a minimum at a temperature-dependent critical field, at temperatures lower than the characteristic temperature. These distinctive features might be possibly associated with magnetic properties of CuO_2 planes arising from an opening of spin gap.

Keywords: intrinsically underdoped magnetoresistance, hybridization, spin gap

二、緣由與目的

It has been of great interest to investigate the physical properties of both normal and superconducting states of underdoped cuprates. NMR[1], neutron[2], and *c*-axis optical conductivity[3] indicate the existence of a pseudogap (or spin gap) in the normal state at a characteristic temperature T^* well above T_c. Photoemission experiments[4-5] suggest that the pseudogap have the same size and ***k*** dependence as the superconducting gap. It has also been found experimentally that T^* increases with a decreasing hole doping δ. Despite considerable theoretical and experimental work, a more complete understanding of the origin of the spin gap is not available.

On the other hand, it was known that all the heavily Pr-doped cuprates, though have identical crystal structures to their relatives that contain rare earth ions, are not superconducting. It is generally believed that the hybridization of the Pr *4f* and its nearest neighbor O $2p_\pi$ orbital, as proposed by

Fehrenbacher and Rice[6], plays a central role in suppressing the superconductivity. The model explains the phenomenon through the localization of holes in hybridized Pr *4f*-O $2p_\pi$ orbitals. In addition to modifying the transport properties of Pr-doped cuprates, Pr *4f*-O $2p_\pi$ hybridization could also be expected to enhance the magnetic coupling between the Pr ions and the CuO_2 planes. Therefore, it is interesting to know how Pr affects the spin dynamics of the CuO_2 planes as well as the magneto-transport of Pr-doped cuprates.

To address this issue, we have systematically studied the temperature dependences of magnetic susceptibility (χ(T)) and magnetoresistance (ΔR/R) of intrinsically underdoped polycrystalline $Y_{1-x}Pr_xBa_2Cu_4O_8$ (x=0.0-1.0) samples. The observed result shows that a characteristic temperature of 180 K, possibly associated with the spin dynamics of CuO_2 planes, hardly changes with the Pr content. It strongly suggests that the hybridization between Pr *4f* and O $2p_\pi$ has only minor effect on the spin-gap related temperature even though the in-plane spin-spin correlation energy may be weakened by the presence of Pr.

The samples investigated were prepared by an oxalate coprecipitation method. The final product was annealed at 500 ℃ for 24 hours in O_2 to get a single-phase compound. Magnetic susceptibility was measured by using a SQUID magnetometer in an applied field of 1000 G. Magnetoresistance and resistivity measurements were performed using a conventional four-probe method. External magnetic fields up to 6 T perpendicular to current direction were applied for MR measurements.

三、結果與討論

Figure 1 shows the temperature dependence of molar magnetic susceptibility χ_M(T) for $YBa_2Cu_4O_8$ (Y124) and $Y_{0.7}Pr_{0.3}Ba_2Cu_4O_8$. As can be seen in Fig. 1, there is a distinct difference between χ_M(T) of Y124 and that of $Y_{0.7}Pr_{0.3}Ba_2Cu_4O_8$. χ_M(T) of Y124 decreases with decreasing temperature, in sharp contrast to the χ_M(T) of a conventional Fermi liquid, which would be temperature independent. There are two regimes in χ_M(T) of Y124 separated by a scale $T_s \sim 180$ K. For $T > T_s$, $\chi_M(T) \cong A + BT$ with $A, B > 0$. For $T < T_s$, χ_M(T) drops more rapidly with a zero extrapolation at $T = 0$. It indicates that there is a reduction in the spin density of states at $T \sim T_s$ associated with the spin dynamics of CuO_2 planes. In contrast to χ_M(T) of Y124, χ_M(T) of Pr-doped compound ($Y_{0.7}Pr_{0.3}Ba_2Cu_4O_8$) exhibits a Curie-Weiss like behavior arising from Pr magnetic moment.

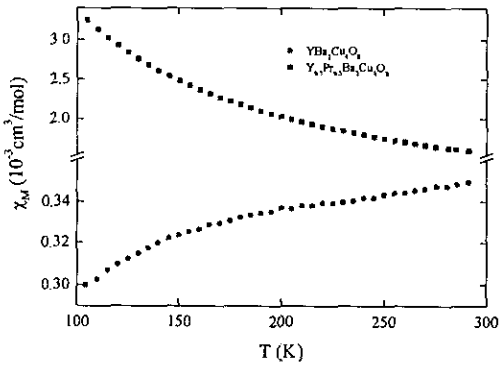

FIG. 1. The temperature dependence of molar magnetic susceptibility χ_M(T) for Y124 and $Y_{0.7}Pr_{0.3}Ba_2Cu_4O_8$.

The presence of a paramagnetic background makes it hard to determine whether a kink around 180 K observed in Y124 still exists in Pr-doped compounds. To further elucidate the nature of this kink as a function of Pr content, inverse of χ_M(T) for $Y_{1-x}Pr_xBa_2Cu_4O_8$ (x =0.3, 06, 0.8, and 1.0) compounds ranging from 100 to 300 K are plotted in Fig. 2.

One remarkable feature of the data displayed in Fig. 2 is that all χ_M^{-1}(T) of the samples investigated can be fitted into two

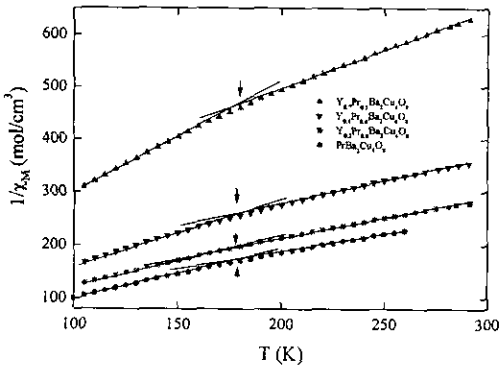

FIG. 2. The inverse of $\chi_M(T)$ for $Y_{1-x}Pr_xBa_2Cu_4O_8$ (x =0.3, 0.6, 0.8, and 1.0) compounds.

straight lines with an interception around 180 K. It should be noted that $\chi(T)$ can be intuitively expressed as $\chi_0(T) + C/T-\theta$, where χ_0 (defined as $\alpha\chi_{Y124}(T)$, α is a fitting parameter) is the contribution from CuO_2 planes and $C/T-\theta$ is the Curie-Weiss term associated with Pr moment. After subtracting the $\alpha\chi_{Y124}(T)$ term, the inverse of $(\chi_M(T)-\alpha\chi_{Y124}(T))$ versus temperature for $Y_{1-x}Pr_xBa_2Cu_4O_{8-\delta}$ (x = 0.0, 0.3, 0.6, 0.8, and 1.0) compounds reveal a linear dependence as shown in Fig. 3.

This plot really illustrates that the remarkable doping-independent kink around 180 K observed in $\chi_M^{-1}(T)$ curve for all Pr-doped samples is associated with the spin dynamics of CuO_2 planes. It appears that the Pr doping doesn't have a significant effect on the spin dynamics of CuO_2 planes. The detailed crystal structure refinement and the Raman scattering measurements show that the oxidation state of Pr ion is 3+ [7] However, the observed effective moment of Pr for $Y_{1-x}Pr_xBa_2Cu_4O_8$ varies from 2.81 μ_B to 2.46 μ_B as x changes from 0.1 to 1.0. It suggests that the hybridization between Pr *4f* and O $2p_\pi$ becomes more pronounced as Pr content increases. The existence of a local Pr *4f* - O $2p_\pi(Pr^{IV})$ hybridized state binds doped holes to Pr sites, which is expected not to affect too much of the hole-doping level. It has been argued that the spin-gap characteristic temperature T_s is correlated with the strength of the in-plane antiferromagnetic fluctuations. As the doping level increases, T_s decreases with the same trend as the antiferromagnetic fluctuation. Therefore, the experimental results of very little effect on the characteristic temperature T_s by increasing Pr content is consistent with such a picture of the strong hybridization between Pr *4f* and O $2p_\pi$ orbitals.

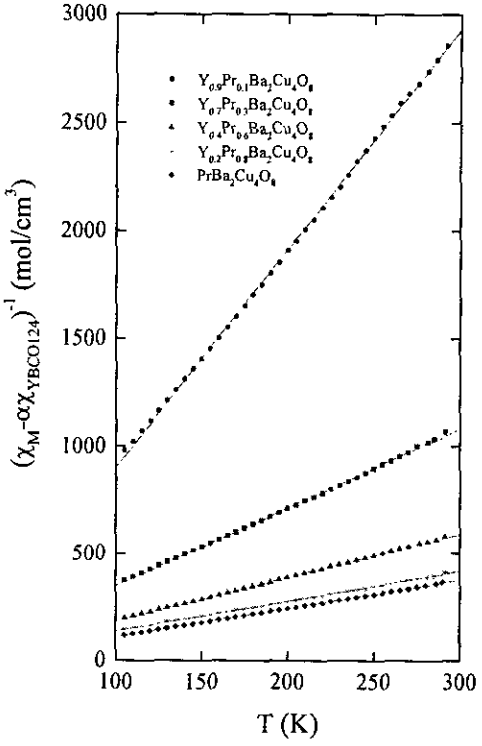

FIG. 3. The inverse of $(\chi_M(T)-\alpha\chi_{Y124}(T))$ versus temperature for $Y_{1-x}Pr_xBa_2Cu_4O_8$ (x= 0.0, 0.3, 0.6, 0.8, and 1.0) compounds.

The magnetic field dependence of the transverse magnetoresistance ratio $\Delta R(H)/R(0)$ of $PrBa_2Cu_4O_8$ (Pr124) at different temperature is shown in Fig. 4. In contrast to the Kohler's scaling rule ($\Delta R/R \propto (H/R)^2$) observed in overdoped cuprates, MR for the Pr124 sample exhibits an intriguing feature below 100 K. As shown in Fig. 4, there is a temperature-dependent minimum around 2 Tesla in $\Delta R/R$ versus H plot. For instance, the MR changes from negative to positive as magnetic field

exceeds 2 Tesla at 4.2 K. More importantly, the negative MR ratio of Pr-doped samples becomes smaller as Pr content decreases, indicating the presence of Pr has significant influence on the negative MR at low temperatures. Another interesting observation is that the transverse MR ratio has comparable order of magnitude with the longitudinal MR ratio. This isotropic MR suggests that a spin part, rather than a orbital part, plays a significant role in the intriguing MR behavior. Furthermore, the observed MR of the Pr124 is distinctly different from Terasaki's results showing only positive MR and roughly proportional to H^2. Quantitatively speaking, $\Delta R(H)/R(0)$ of observed data can be expressed as $a(T)H + b(T)H^{1.4}$ with $a < 0$ and $b > 0$. The fitting parameters a and b become vanishingly small as temperature approaches 100 K, suggesting the striking MR feature might be closely related to the fact that R(T) of Pr124 is proportional to $T^{1.5}$ below 100 K as well as the superconducting state in the CuO_2 planes.

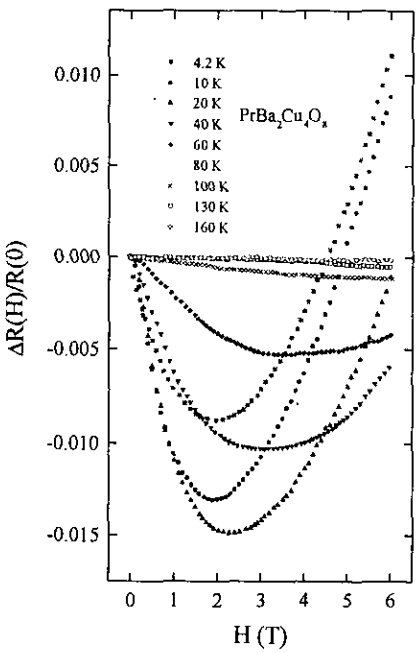

FIG. 4 The magnetic field dependence of transverse MR ratio of Pr124 at different temperatures.

It should be noted that the largest negative MR ratio was observed around 10~20 K, indicating that Pr ordering at 17 K might be responsible for the negative MR in Pr-doped samples. A possible scenario for the striking MR behavior is the following. In lower filed regime, field suppresses the spin fluctuation in the CuO_2 planes and reduces scattering rate between preformed bosons and carriers. It gives rise to a negative MR. Whereas in higher field regime, field just changes the trajectory of carriers to cause a positive MR.

四、計劃成果自評

In summary, our detailed systematic magnetic studies on the $Y_{1-x}Pr_xBa_2Cu_4O_8$ system indicate that the suppression of superconductivity by Pr-doping is closely associated with the strong hybridization between Pr *4f* and O $2p_\pi$ orbital, which consequently causes the localization of mobile holes. However, this hybridization has only minor effect on the spin dynamics, which can be characterized by the presence of a characteristic spin gap temperature, of the cuprates. We certainly believe that the obtained results really shine a light on understanding of the normal state properties of underdoped high-T_c cuprates as well as underlying mechanism of the spin gap.

五、參考文獻

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