Possible ferromagnetic phase in non-superconducting heavily overdoped cuprates of Bi-2201

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Abstract. In order to investigate the possible ferromagnetic phase in the non-superconducting (non-SC) heavily overdoped (HOD) regime of high-T_c cuprates suggested by Sonier *et al.* [Proc. Natl. Acad. Sci. USA **107**, 17131 (2010)] in $La_{2-x}Sr_xCuO_4$, both the magnetization and in-plane electrical resistivity, ρ_{ab}, have been measured using single crystals of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ covering a wide range of hole concentration. Magnetization curves in the non-SC HOD regime have shown a tendency to be saturated in high magnetic fields at low temperatures, which is probably a precursor of the possible ferromagnetic transition at a lower temperature. In addition, it has been found that ρ_{ab} exhibits the temperature dependence being proportional to $T^{4/3}$, which is characteristic of a metal with two-dimensional ferromagnetic fluctuations, in a wide range of hole concentration from the superconducting overdoped to the non-SC HOD regime. It has been concluded that two-dimensional ferromagnetic correlation develops at low temperatures in HOD Bi-2201 and that therefore the development of the ferromagnetic correlation is probably an intrinsic feature in the HOD cuprates.

1. Introduction

In general, electrons in the non-superconducting (non-SC) heavily overdoped (HOD) regime of high-T_c cuprates have been believed to behave as Fermi-liquid. However, phenomena contradictory to the Fermi-liquid picture have been reported, such as an upturn of the magnetic susceptibility, χ, at low temperatures in HOD $Tl_2Ba_2CuO_{6+\delta}$ [1]. Formerly, it has theoretically been suggested that ferromagnetic fluctuations suppress the *d*-wave superconductivity in the overdoped (OD) regime of hole-doped high-T_c cuprates [2]. In addition, Sonier *et al.* have reported from muon spin relaxation measurements in non-SC HOD $La_{2-x}Sr_xCuO_4$ (LSCO) with $x = 0.33$ that the spin correlation develops at low temperatures below 0.9 K and that the in-plane electrical resistivity, ρ_{ab}, is proportional to $T^{5/3}$ at high temperatures above 60 K, suggesting the existence of three-dimensional (3D) ferromagnetic fluctuations in the non-SC HOD regime [3]. However, both oxygen deficiency and disorder characteristic of non-SC HOD LSCO give us a doubt whether a ferromagnetic phase intrinsically exists in the cuprates.

The so-called Bi-2201 is one of promising high-T_c cuprates in which the partial substitution of Pb for Bi allows us to access to the non-SC HOD regime. Moreover, Bi-2201 is free from oxygen deficiency and expected to be less disordered than LSCO. Therefore, in order to investigate the possible ferromagnetic phase, we have performed both magnetization and ρ_{ab} measurements using Bi-2201 single crystals of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ in the superconducting (SC) regime and non-SC OD regime.

2. Experimental

$Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ single crystals were grown by the floating-zone technique. The quality of the crystals obtained was evaluated to be good from the observation of clear fourfold symmetric spots in the x-ray back-Laue photograph. The composition of the crystals was determined by the inductively-coupled-plasma analysis. To control the hole concentration, reduction or oxidation annealing was performed in vacuum, in argon or in oxygen of ambient or high pressure [4-6]. The magnetization was measured using a SQUID magnetometer. The ρ_{ab} was measured by the standard dc four-probe method using a commercial apparatus (Quantum Design, PPMS). The value of the hole concentration, p, in the SC crystals was determined using the empirical law of $T_c/T_c^{max} = 1 - 82.6(p[\text{per Cu}] - 0.16)^2$, where T_c^{max} is the maximum value of T_c in a system [7]. In this study, four kinds of crystals were used; HOD1 and HOD2 in the non-SC HOD regime with $p > 0.27$ per Cu, and OD1 and OD2 in the SC overdoped regime with p = 0.26, 0.22 per Cu and T_c = 2.8, 15.0 K, respectively.

3. Results and discussion

Figure 1 shows the temperature dependence of χ for crystals annealed under various conditions. The χ is given by

$$\chi(T) = \chi_0 + \chi_1(T), \tag{1}$$

where χ_0 and $\chi_1(T)$ are temperature-independent and temperature-dependent terms, respectively. The χ_0 is described as

$$\chi_0 = \chi_{\text{Pauli}} + \chi_{\text{core}} + \chi_{\text{V.V.}}, \tag{2}$$

where χ_{Pauli} is the Pauli paramagnetism of conducting electrons, χ_{core} the core diamagnetism and $\chi_{\text{V.V.}}$ the Van Vleck paramagnetism. Since only χ_{Pauli} among them depends on p and increases with increasing p, the magnitude relation of p between the crystals can be estimated roughly from the value of $\chi(T)$ at high temperatures. In fact, $\chi(T)$ of the crystal OD1 with T_c = 2.8 K is larger than that of the crystal OD2 with T_c = 14.8 K. The p value of the non-SC crystal HOD2 is estimated to be larger than that of the crystal OD1 and that of the crystal HOD1 is estimated to be larger than that of the crystal HOD2. In Fig. 1, it is also found that χ at low temperatures exhibits an upturn in crystals HOD1, HOD2 and OD1, as observed in HOD $Tl_2Ba_2CuO_{6+\delta}$ [1]. This may be due to ferromagnetic fluctuations.

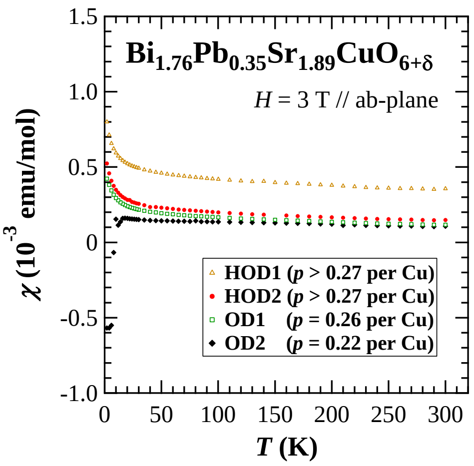

Figure 1. Temperature dependence of the magnetic susceptibility, χ, of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ in the overdoped (OD) and the heavily overdoped (HOD) regimes.

Figure 2(a) shows the magnetization curves of the crystal HOD2 in the HOD regime. Surprisingly, while the magnetization is linear to magnetic field at 20 K, it tends to be saturated in high fields at low

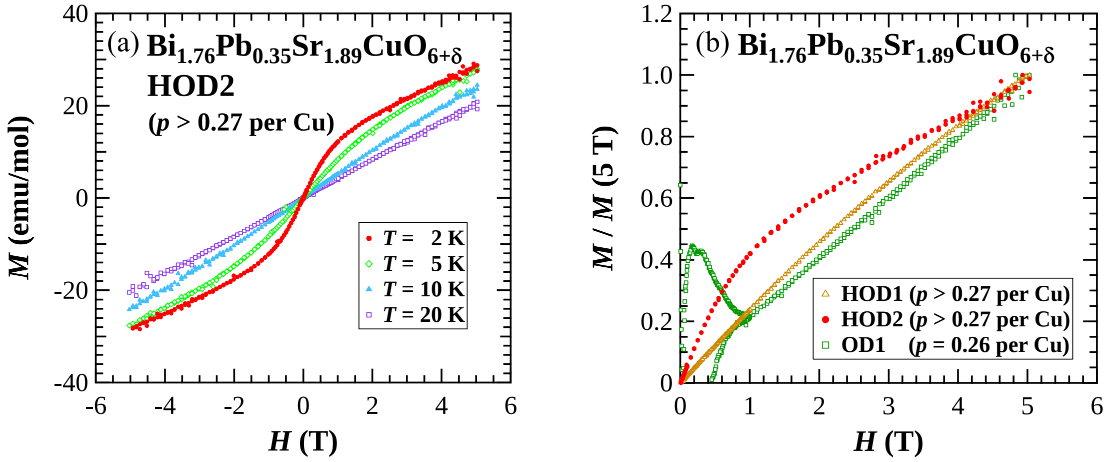

Figure 2. (a) Magnetization curves of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ in the heavily overdoped (HOD) regime. (b) Magnetization curves at 2 K of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ in the overdoped (OD) and HOD regimes normalized by values of magnetization at 5 T, M/M(5 T).

temperatures. This is probably a precursor of the ferromagnetic transition pointed out by Sonier *et al.* in non-SC HOD LSCO with $x = 0.33$ [3]. In Fig. 2(b), the normalized magnetization curve at 2 K is shown for crystals OD1, HOD1 and HOD2. It is clearly found that the tendency to the saturation is observed only in the HOD regime and that the saturation becomes more significant in the crystal HOD2 close to the SC regime than the crystal HOD1. Therefore, it appears that the ferromagnetic transition takes place at a low temperature below 2 K in the HOD regime and the transition temperature is the highest at the boundary between SC OD and non-SC HOD regimes.

Figure 3(a) shows the temperature dependence of ρ_{ab} of crystals OD1 and HOD1. It is found that ρ_{ab} is proportional to $T^{4/3}$ in a wide range of temperature. The p dependence of the exponent n obtained by the fitting with $\rho_{ab} = \rho_0 + AT^n$ in the normal state is shown in Fig. 3(b). It is found that the n value changes from 1 around the optimally doped regime to 2 in the underdoped regime, as pointed out by

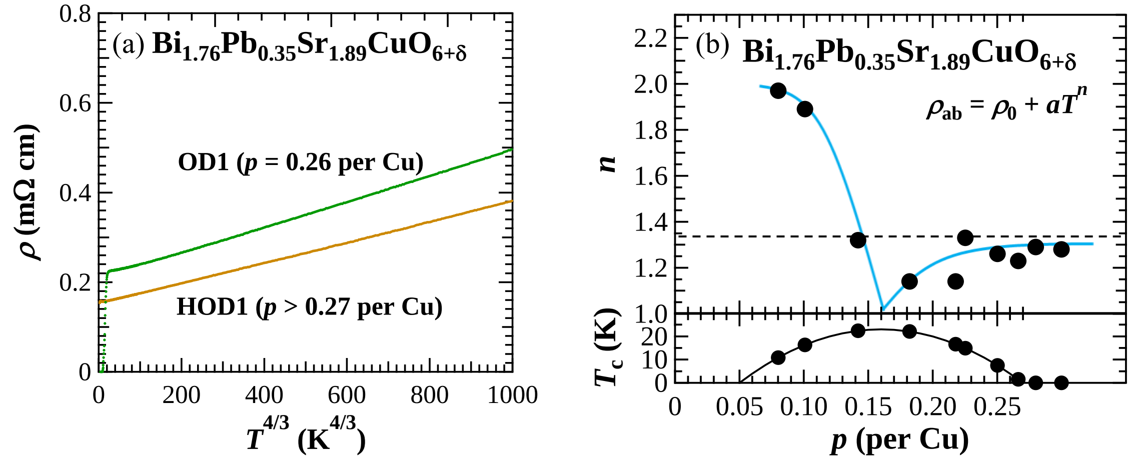

Figure 3. (a)Temperature dependence of the in-plane electrical resistivity, ρ_{ab}, of $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ in the overdoped (OD) and heavily overdoped (HOD) regimes. (b) Hole-concentration, p, dependences of the exponent n in $\rho_{ab} = \rho_0 + AT^n$ and T_c in $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$. The broken line exhibits $n = 4/3$. The solid line in n vs. p is a guide line and the solid line in T_c vs. p is given by the empirical relation [7].

Ando *et al*. [8]. On the other hand, from the optimally doped regime to the HOD regime, the n value tends to increase with increasing p and to converge to 4/3 in the HOD regime. According to the self-consistent renormalization theory [9], the $T^{4/3}$ dependence of ρ_{ab} is characteristic of a metal with two-dimensional (2D) ferromagnetic fluctuations. This result is associated with the $T^{5/3}$ dependence of ρ_{ab} observed in non-SC HOD LSCO, which is characteristic of 3D ferromagnetic fluctuations [3]. The difference in dimensionality may be caused by the weakness of the magnetic coupling between CuO_2 layers due to the long interlayer distance of Bi-2201 (12.3 Å) compared with that of LSCO (6.6 Å).

From the theoretical point of view, it has been suggested that a ferromagnetic order can be formed in a metal where the electron correlation is strong enough, using the mean field approximation [10] and the Gutzwiller approximation [11], but Tahvildar-Zadeh *et al*. have reported that the ferromagnetic correlation will be collapsed by quantum fluctuations within the 3D Hubbard model [12]. Experimentally, Piriou *et al*. have recently reported that the density of states increases with increasing p in the HOD regime due to the existence of the van Hove singularity from the scanning tunneling spectroscopy in Bi-2201 single crystals [13]. If this is the case, it is possible that a ferromagnetic order appears, satisfying the Stoner's criterion. Therefore, the present results of the magnetization and ρ_{ab} suggesting the existence of ferromagnetic fluctuations may be due to the enhancement of the density of states at the Fermi level owing to the approach to the van Hove singularity in the HOD regime and a ferromagnetic transition is expected to take place at a low temperature below 2 K.

4. Summary

We have investigated the possible ferromagnetic state in the HOD regime of high-T_c cuprates using $Bi_{1.76}Pb_{0.35}Sr_{1.89}CuO_{6+\delta}$ single crystals covering a wide range of p. It has been found that χ exhibits an upturn at low temperatures in the OD and HOD regimes and that the magnetization curves exhibit a tendency to be saturated in high fields at low temperatures in the non-SC HOD regime, which is the most significant at p close to the SC regime. Moreover, $T^{4/3}$ dependence of ρ_{ab} has been observed in the OD and HOD regimes, suggesting the existence of 2D ferromagnetic fluctuations. It has been concluded that 2D ferromagnetic correlation develops at low temperatures in the HOD regime of Bi-2201, so that a ferromagnetic transition is expected to take place at a lower temperature below 2 K. Accordingly, the development of the ferromagnetic correlation might be an intrinsic feature in the HOD cuprates.

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