Magnetic susceptibility of optimally doped $HgBa_2CuO_{4+\delta}$

Yutaka Itoh,[1,*] Takato Machi,[2] and Ayako Yamamoto[3]

[1]*Department of Physics, Graduate School of Science, Kyoto Sangyo University, Kamigamo-Motoyama, Kika-ku, Kyoto 603-8555, Japan*

[2]*AIST Tsukuba East, Research Institute for Energy Conservation, 1-2-1 Namiki, Tsukuba, Ibaraki 305-8564, Japan*

[3]*Graduate School of Engineering and Science, Shibaura Institute of Technology, 3-7-5 Toyosu, Koto-ku, Tokyo 135-8548, Japan*

(Received 29 August 2017; revised manuscript received 13 November 2017; published 13 December 2017)

The magnitude of the powder spin susceptibility of an optimally doped superconductor $HgBa_2CuO_{4+\delta}$ (Hg1201) in the normal state is found to be nearly the same as that of $La_{2-x}Sr_xCuO_4$ near the optimally doped level. The Stoner enhancement factor of Hg1201 is larger than that of $La_{2-x}Sr_xCuO_4$. The magnitude correlation of the Stoner enhancement factor is inconsistent with the effect of the recent theoretical Coulomb repulsion between $3d$ electrons and that of the superexchange interaction of a charge transfer type.

DOI: 10.1103/PhysRevB.96.235118

I. INTRODUCTION

Uniform spin susceptibility χ_s is a fundamental property of strongly correlated electron systems to understand the many-body effects. It is the $\vec{q} = 0$ component of the static spin susceptibility $\chi_s'(\vec{q})$ ($\vec{q}$ is the wave vector). For typical high-T_c cuprate superconductors in the underdoped regime, the normal state χ_s decreases with decreasing temperature, which is known to be the pseudogap effect. In the optimally doped regime, χ_s is nearly independent of temperature. In the overdoped regime, χ_s is still nearly independent of temperature, increases with decreasing temperature, or makes a broad maximum above T_c. The magnitude of the temperature-independent χ_s also tells us how different it is from the conventional Fermi-liquid theory and how much is the degree of the Coulomb repulsion effect through the Stoner exchange enhancement factor. Recent theoretical calculations of the Coulomb repulsion U from first principles indicate that the lower T_c $La_{2-x}Sr_xCuO_4$ has stronger U than $HgBa_2CuO_{4+\delta}$ (Hg1201) by 1.47 times [1]. The estimation of the Stoner factor could be an experimental test of the Coulomb repulsion U in the itinerant systems.

In this paper, we report on the measurement and analysis of the bulk magnetic susceptibility of a single-CuO_2-layer high-T_c superconductor Hg1201 at the optimally doped level ($T_c = 98$ K) in the normal state. We found that the Stoner enhancement factor of Hg1201 is larger than that of $La_{2-x}Sr_xCuO_4$, which is in contrast to the recent theoretical calculations of U. Discussions were made from a single-band Hubbard model and a t-J model within the random-phase approximation.

II. EXPERIMENTS

High-quality polycrystalline samples of Hg1201 were prepared by a solid-state reaction with high-purity BaO powder, which was a key to heat treatment at a relatively high temperature of 930 °C [2]. The sample for the present study has been confirmed to be in a single phase by powder x-ray diffraction patterns and characterized by transport measurements [2]. The dc magnetic susceptibility χ at an external magnetic field of 1.0 T was measured by a superconducting quantum interference device (SQUID) magnetometer (QUANTUM Design, MPMS). From the previous magnetization measurement in a field of 20 Oe, the superconducting transition temperature T_c was estimated to be 98 K [2]. For the comparative studies on the powder spin susceptibility, the polycrystalline samples of $La_{2-x}Sr_xCuO_4$ ($x = 0.13$, 0.15, and 0.18) were synthesized by a conventional solid-state reaction method [3]. After they were confirmed to be of a single phase by the powder x-ray diffraction patterns, their powder magnetic susceptibilities were measured by the SQUID magnetometer. The samples of $x = 0.13$, 0.15, and 0.18 exhibit $T_c = 34$, 38, and 35 K, respectively.

The powder susceptibility χ is the isotropic part of the magnetic susceptibility $(\chi_{aa} + \chi_{bb} + \chi_{cc})/3$ ($\chi_{\alpha\alpha}$ in a field along the α axis). The anisotropic part of the magnetic susceptibility is known to be due to the Van Vleck orbital susceptibility [4,5]. Although the recently grown single crystals [6] will enable us to obtain the tensor components in $\chi_{\alpha\alpha}$, the powder magnetic susceptibility can tell us the isotropic part of the spin susceptibility and how much the electron correlation effect is.

III. EXPERIMENTAL RESULTS

A. Spin susceptibility of Hg1201

Figure 1 shows the magnetic susceptibility χ of the powder Hg1201. χ levels off above about 250 K and decreases with cooling to $T_c = 98$ K, which is a pseudogap behavior. Since the temperature dependence of χ resembles those of the plane-site ^{63}Cu and ^{17}O Knight shifts [7–9], one may regard the present bulk magnetic susceptibility χ as an intrinsic behavior.

The bulk magnetic susceptibility is the sum of the individual components, $\chi = \chi_s + \chi_{vv} + \chi_{dia}$, where χ_s is the spin susceptibility of the $3d$ electrons, χ_{vv} is the powder-averaged Van Vleck orbital susceptibility ($+0.23 \times 10^{-4}$ emu/mole-f.u. after the band calculation for Sc_2CuO_4 [10] and according to the analysis for $La_{2-x}Sr_xCuO_4$ [5,11]), and χ_{dia} is the diamagnetic susceptibility of the inner-shell electrons in the core (-1.44×10^{-4} emu/mole-f.u. [12]). The spin susceptibility χ_s is obtained from $\chi_s = \chi - \chi_{vv} - \chi_{dia}$. The estimated $\chi_s = 1.47 \times 10^{-4}$ emu/mole-f.u. of Hg1201 ($T > 250$ K) in Fig. 1 is nearly the same as the reported $\chi_s = 1.4$–1.7×10^{-4}

*yitoh@cc.kyoto-su.ac.jp

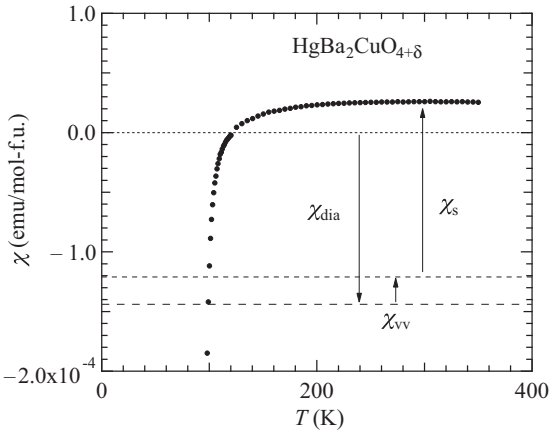

FIG. 1. Bulk magnetic susceptibility χ of powdered $HgBa_2CuO_{4+\delta}$ ($T_c \approx 98$ K) in an external magnetic field of 1.0 T. The bulk χ is the sum of inner-core electron diamagnetic susceptibility $\chi_{dia}(<0)$, Van Vleck orbital susceptibility $\chi_{vv}(>0)$, and $3d$ electron spin susceptibility χ_s.

emu/mole-f.u. of $La_{2-x}Sr_xCuO_4$ near the optimally doped level [11,13–15].

Figure 2 shows the spin susceptibility $\chi_s/N_A\mu_B^2$ of an optimally doped Hg1201, where N_A is Avogadro's number and μ_B is the Bohr magneton. The solid and dashed lines indicate the bare spin susceptibility $\chi_0/N_A\mu_B^2 = N(E_F)$ from the band theoretical calculation [16], where $N(E_F)$ is the electron density of states at the Fermi energy E_F for both spin directions in units of states/eV-f.u. and the electron g factor is assumed to be 2.

B. Hg1201 vs $La_{2-x}Sr_xCuO_4$

Figure 3(a) shows the powder magnetic susceptibilities χ in emu/g of $La_{2-x}Sr_xCuO_4$ ($x = 0.13, 0.15, 0.18$) in an external magnetic field of 1.0 T for comparison with Hg1201. The magnitude of χ of $La_{2-x}Sr_xCuO_4$ is nearly the same as those in the literature [11,13–15].

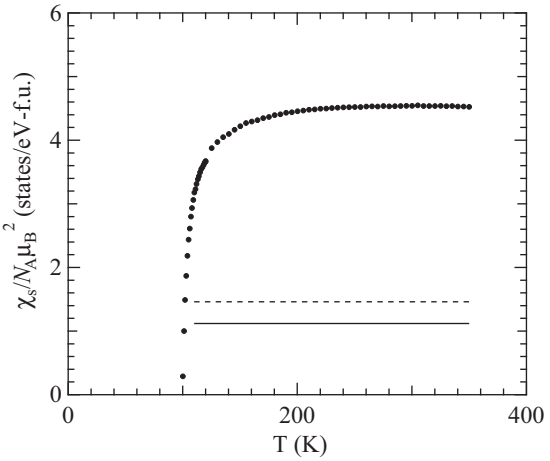

FIG. 2. Uniform spin susceptibility $\chi_s/N_A\mu_B^2$ in states/eV-f.u. for $HgBa_2CuO_{4+\delta}$ ($T_c \approx 98$ K). The solid and dashed lines are the electron density of states from the band theoretical calculations [16].

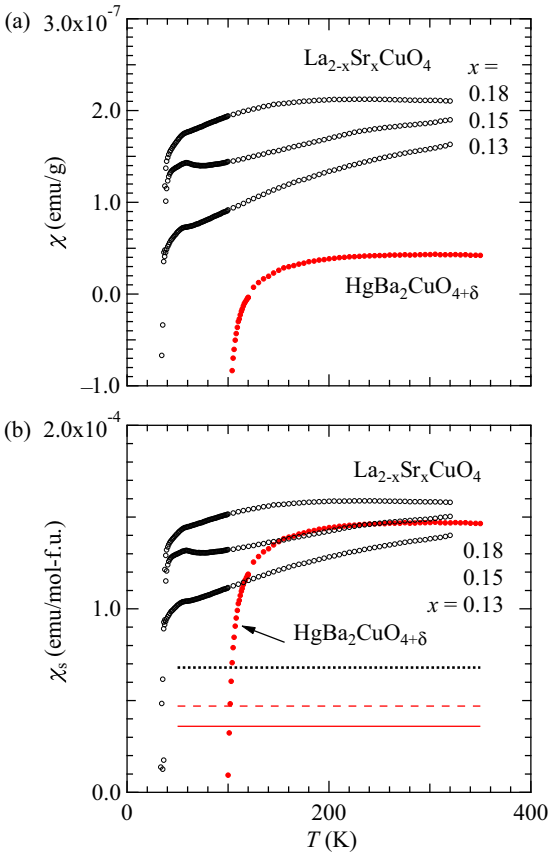

FIG. 3. (a) Powder magnetic susceptibilities χ in emu/g of optimally doped Hg1201 and $La_{2-x}Sr_xCuO_4$ ($x = 0.13, 0.15, 0.18$) in an external magnetic field of 1.0 T. (b) Powder spin susceptibilities χ_s in emu/mol-f.u. of optimally doped Hg1201 and $La_{2-x}Sr_xCuO_4$ ($x = 0.13, 0.15, 0.18$). The dotted line is the band theoretical spin susceptibility χ_0 for $La_{2-x}Sr_xCuO_4$ [17,18]. The solid and dashed lines are the band theoretical spin susceptibility χ_0 for Hg1201 [16].

Figure 3(b) shows the powder spin susceptibilities χ_s in emu/mol-f.u. of $La_{2-x}Sr_xCuO_4$, which are obtained from $\chi_s = \chi - \chi_{vv} - \chi_{dia}$ in the same procedure of the spin-orbital partition as Hg1201. The orbital susceptibility χ_{vv} is taken to be $+0.23 \times 10^{-4}$ emu/mole-f.u. [5,10,11]. The core electron susceptibility χ_{dia} with the Sr concentration x is taken to be $(-99 + 5x) \times 10^{-6}$ emu/mole-f.u. [12].

In Fig. 3(b), the magnitude of the spin susceptibility χ_s of Hg1201 above 250 K is nearly the same as those of $La_{2-x}Sr_xCuO_4$ for $x = 0.13, 0.15, 0.18$. The band theoretical calculations of the spin susceptibility χ_0 are shown by a dotted line for $La_{2-x}Sr_xCuO_4$ [17,18] and by solid and dashed lines for Hg1201 [16].

The band theoretical $N(E_F)$ of $La_{1.85}Sr_{0.15}CuO_4$ [17,18] is larger than that of Hg1201 [16] by about 1.4–1.8 times, while the experimental χ_s of $La_{1.85}Sr_{0.15}CuO_4$ is estimated to be nearly the same as that of Hg1201. For Hg1201, we estimated the Stoner exchange enhancement factor $S = \chi_s/\chi_0$ $(= 1/[1 - IN(E_F)]$ in the random phase approximation for an effective interaction I) to be 4.1 at the moderately doped level and 3.2 at the two-dimensional van Hove singularity. The smaller orbital susceptibility $\chi_{vv} = +0.15 \times 10^{-4}$ emu/mole-

TABLE I. Electron density of states $N(E_F)$ from band theoretical calculations [16–18], experimental spin susceptibilities χ_s for $La_{1.8}Sr_{0.2}CuO_4$ [11] and the present Hg1201 ($T > 250$ K), Stoner enhancement factors S, and effective interactions I. $N(E_F)$ and $\chi_s/N_A\mu_B^2$ are shown in state/eV formula units, and I in eV.

	$N(E_F)$	$\chi_s/N_A\mu_B^2$	S	I
$La_{1.8}Sr_{0.2}CuO_4$	2.09	4.3	2.1	0.25
$HgBa_2CuO_{4+\delta}$	1.46	4.5	3.2	0.47
	1.12	4.5	4.1	0.68

f.u. estimated experimentally in [15] leads to more enhanced S in χ_s. The Stoner enhancement factor S of Hg1201 is 1.5–2.0 times larger than $S \sim 2$ of $La_{1.8}Sr_{0.2}CuO_4$ [11] and $S =$ 2.0–2.3 (320 K) of the present $La_{2-x}Sr_xCuO_4$ ($x = 0.13, 0.15,$ 0.18) in Fig. 3(b). The effective interaction I of Hg1201 is stronger than that of $La_{2-x}Sr_xCuO_4$, I(Hg1201) $>$ I(LSCO). The estimated parameters are shown in Table I.

IV. DISCUSSIONS

The recent first-principles calculations indicate that the on-site Coulomb repulsion U between $3d$ electrons in Hg1201 is weaker than that in $La_{2-x}Sr_xCuO_4$ by 0.68 times [1]. The value of $U = 3.15$ eV in La_2CuO_4 is calculated to be larger than $U =$ 2.15 eV in Hg1201 [1]. In the two-dimensional Hubbard model with the random-phase approximation, the effective interaction I in the Stoner factor is an effective Coulomb repulsion $\bar{U}$ [19],

$$\chi_s = \frac{\chi_0}{1 - \bar{U}N(E_F)}. \quad (1)$$

The band theories indicate $N(E_F)$ of Hg1201 to be smaller than that of $La_{2-x}Sr_xCuO_4$ in Table I [16–18]. If one assumes that the magnitude correlation on $\bar{U}$ between Hg1201 and $La_{2-x}Sr_xCuO_4$ (LSCO) is the same as U, that is, $\bar{U}$(Hg1201) $<$ $\bar{U}$(LSCO), then the Stoner enhancement factor of Hg1201 must be smaller than that of $La_{2-x}Sr_xCuO_4$. The present experimental estimation of the Stoner enhancement factor of Hg1201 that is larger than that of $La_{2-x}Sr_xCuO_4$ is in contrast to the effect of the theoretical Coulomb repulsion U.

In the two-dimensional t-J model with the random-phase approximation, the effective interaction I in the Stoner factor corresponds to a superexchange interaction J_0 [20],

$$\chi_s = \frac{\chi_0}{1 + J_0N(E_F)}. \quad (2)$$

The superexchange interaction J_0 of a charge transfer type is expressed as

$$J_0 \propto \frac{4T_{pd\sigma}^4}{\Delta_{ct}^2}\left(\frac{1}{\Delta_{ct}} + \frac{1}{U}\right), \quad (3)$$

where $T_{pd\sigma}$ is a p-d hybridization matrix element and Δ_{ct} is a charge transfer gap [21]. The value of $T_{pd\sigma}$ in La_2CuO_4 is nearly the same as that in Hg1201 [1]. The value of the d-p charge transfer energy $\Delta_{dp} = 2.58$ eV in La_2CuO_4 is larger than $\Delta_{dp} = 1.84$ eV in Hg1201 [1]. According to Eq. (3), larger U and $\Delta_{ct}(\propto \Delta_{dp})$ in $La_{2-x}Sr_xCuO_4$ than in Hg1201 lead to smaller J_0 in $La_{2-x}Sr_xCuO_4$ than in Hg1201 [J_0(Hg1201) $>$ J_0(LSCO)]. The Stoner enhancement factor in Eq. (2) for Hg1201 is smaller than that for $La_{2-x}Sr_xCuO_4$, which is also in contrast to the experimental magnitude correlation in the Stoner factor. No parent antiferromagnetic insulator has been found in Hg1201, perhaps due to chemical instability. It could not be tested whether the magnitude of J_0 is larger in the Mott insulating state of Hg1201 than that of La_2CuO_4.

The single-CuO_2-layer superconductor $Tl_2Ba_2CuO_{6+\delta}$ ($T_c = 85$ K) also shows a large Stoner enhancement factor, $S = 3.8$–$4.2 \sim 4$ [22], which is comparable to the present Hg1201 ($T_c = 98$ K) in Table I. The large Stoner enhancement factor may characterize the higher-T_c superconductors more than $La_{2-x}Sr_xCuO_4$ and $Bi_2Sr_2CuO_{6+\delta}$ [23].

V. CONCLUSION

In conclusion, the magnitude of the uniform spin susceptibility of the optimally doped Hg1201 is nearly the same as that of $La_{2-x}Sr_xCuO_4$. The Stoner enhancement factor of Hg1201 is larger than that of $La_{2-x}Sr_xCuO_4$. The effective interaction I is in contrast to the recent first-principles calculations on the Coulomb repulsion U and the effect of the superexchange interaction J_0 in the t-J model within the random-phase approximation.

[1] S. W. Jang, H. Sakakibara, H. Kino, T. Kotani, K. Kuroki, and M. J. Han, Sci. Rep. **6**, 33397 (2016).

[2] A. Yamamoto, W.-Z. Hu, and S. Tajima, Phys. Rev. B **63**, 024504 (2000).

[3] P. G. Radaelli, D. G. Hinks, A. W. Mitchell, B. A. Hunter, J. L. Wagner, B. Dabrowski, K. G. Vandervoort, H. K. Viswanathan, and J. D. Jorgensen, Phys. Rev. B **49**, 4163 (1994).

[4] T. Shimizu, H. Aoki, H. Yasuoka, T. Tsuda, Y. Ueda, K. Yoshimura, and K. Kosuge, J. Phys. Soc. Jpn. **62**, 3710 (1993).

[5] R. E. Walstedt and S.-W. Cheong, Phys. Rev. B **64**, 014404 (2001).

[6] X. Zhao, G. Yu, Y.-C. Cho, G. Chabot-Couture, N. Barišić, P. Bourges, N. Kaneko, Y. Li, L. Lu, M. E. M. Motoyama, O. P. Vajk, and M. Greven, Adv. Mater. **18**, 3243 (2006).

[7] J. Bobroff, H. Alloul, P. Mendels, V. Viallet, J.-F. Marucco, and D. Colson, Phys. Rev. Lett. **78**, 3757 (1997).

[8] Y. Itoh, T. Machi, S. Adachi, A. Fukuoka, K. Tanabe, and H. Yasuoka, J. Phys. Soc. Jpn. **67**, 312 (1998).

[9] D. Rybicki, J. Kohlrautz, J. Haase, M. Greven, X. Zhao, M. K. Chan, C. J. Dorow, and M. J. Veit, Phys. Rev. B **92**, 081115 (2015).

[10] T. C. Leung, X. W. Wang, and B. N. Harmon, Phys. Rev. B **37**, 384 (1988).

[11] D. C. Johnston, Phys. Rev. Lett. **62**, 957 (1989).

[12] *Landolt-Börnstein New Series, Group II*, edited by K. H.

Hellwege and A. M. Hellwege (Springer-Verlag, Heidelberg, 1986), Vol. 16.
[13] L. F. Schneemeyer, J. V. Waszczak, E. A. Rietman, and R. J. Cava, Phys. Rev. B **35**, 8421 (1987).
[14] I. Terasaki, M. Hase, A. Maeda, K. Uchinokura, T. Kimura, K. Kishio, I. Tanaka, and H. Kojima, Physica C **193**, 365 (1992).
[15] M. Hücker, G. D. Gu, and J. M. Tranquada, Phys. Rev. B **78**, 214507 (2008).
[16] D. L. Novikov and A. J. Freeman, Physica C **212**, 233 (1993).
[17] P. B. Allen, W. E. Pickett, and H. Krakauer, Phys. Rev. B **36**, 3926 (1987).
[18] J.-H. Xu, T. J. Watson-Yang, J. Yu, and A. J. Freeman, Phys. Lett. A **120**, 489 (1987).
[19] N. Bulut, D. W. Hone, D. J. Scalapino, and N. E. Bickers, Phys. Rev. B **41**, 1797 (1990).
[20] T. Tanamoto, H. Kohno, and H. Fukuyama, J. Phys. Soc. Jpn. **62**, 717 (1993).
[21] J. Zaanen and G. A. Sawatzky, Can. J. Phys. **65**, 1262 (1987).
[22] D. J. Singh and W. E. Pickett, Physica C **203**, 193 (1992).
[23] G. Le Bras, Z. Konstantinovic, D. Colson, A. Forget, J.-P. Carton, C. Ayache, F. Jean, G. Collin, and Y. Dumont, Phys. Rev. B **66**, 174517 (2002).