TEMPERATURE-LINEAR TERM OF HEAT CAPACITY OF La-Ba-Cu-O AND La-Sr-Cu-O SYSTEMS

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Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

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Abstract. – Heat capacity of $La_{2-x}M_xCuO_4$ (M = Ba, Sr) has been investigated between 1.5 and 10 K. The temperatureelinear term, γT, of heat capacity is absent in the antiferromagnetic ordered region of $x < 0.02$. γ increases abruptly from $x = 0.02$ to $x = 0.05$ where the system is insulating at low temperature and reaches of the order of 4mJ/mol.K^2. The appearance of the finite γ between the AF and SC state causes clearly to spin degrees of freedom. In the superconducting region, γ changes strongly with x and seems to be vanishingly small at $x = 0.10$ for Ba-doping and 0<x<0.20 for Sr-doping.

High-T_c superconductors show a rich phase diagram involving structural, electronic (insulator-metal transition) and magnetic phase transitions. Figure 1 represents the phase diagram of La-Ba-Cu-O which summarize the known experiments [1]. The fundamental understanding of the mechanism of the high-T_c superconductivity remains still unresolved up to now. Anderson proposed a striking theory with a new concept on the physics, which predicted the finite γ of heat capacity in the superconducting (SC) state due to a gapless spinons excitation spectrum [2]. Experimentally, finite γT term in low temperature heat capacity were observed in both La and Y systems by many groups [3, 4].

Recent systematic studies of low temperature heat capacity of La system show the strong correlation between vanishing γT in the antiferromagnetic (AF) state and finite γT term in an insulating state for Ba- and Sr-doping [5, 6]. Detailed doping dependence of γ in the SC state must be urgently investigated because of poor reproducibility of γ values in the SC state in the early studies. In this paper, we report the strong variation of γ with Ba- and Sr-doping in the both insulating and SC state. The major result is that γ in the SC state is vanishingly small at $x = 0.10$ for Ba-doping and $0.10 < x < 0.20$ for Sr-doping.

Samples were prepared by solid state reaction of a

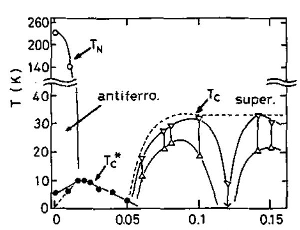

Fig. 1. – Phase diagram of $La_{2-x}Ba_xCuO_4$. Open circles are determined by the internal field analysis, and closed circles are obtained from the relaxation anomalies [7]. Superconducting transtion temperatures are determined by the onset (∇) and midpoint (Δ) of transition of ac susceptibility. Doted line shows the onset temperatures of electrical resistance.

mixture of La_2O_3, $BaCO_3$, $SrCO_3$ and CuO. Details for sample preparations are reported elsewhere [5, 7]. Heat capacity was measured by heat-pulsed method with calibrated Ge thermometer between 1.5 and 10 K.

Figure 2 shows heat capacity for Ba- doped samples by plots of $C\ /\ T$ *vs.* T^2. The temperature dependence of heat capacity is fitted by the $C = \gamma T + \beta T^3$ relation in the low temperature region. Here, T^3 term is from the phonon contribution in the Debye model.

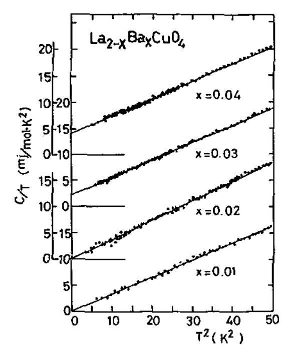

Fig. 2. – Heat capacity plottes as C/T *vs.* T^2 for Ba-doped samples. The solid lines are obtained by least square fitting in the low temperature region.

The magnitude of the T-linear term is obtained from extrapolating to $T = 0$ K by least square fitting. The concentration dependence of γ is shown in figure 3. As reported already in the Ba- and Sr-doped systems [5, 6], it should be emphasized that γ is absent in the AF region and increases when 3D-AF order disappears. Thus, our data indicate a fairly strong correlation between magnetic order and vanishingly small γT term and suggests an intrinsic origin for the γT term when it is finite. In the range of $x < 0.06$, we have confirmed the anomalies of nuclear relaxation rates at low temperatures [7]. The relaxation anomalies, which is clearly associated with the drastic change of fluctuations of spins in the system, is correlated with the large contribution to heat capacity in the insulating state.

In the SC state of $x > 0.06$, we have unexpectedly dramatic x-dependence of the γ values, although

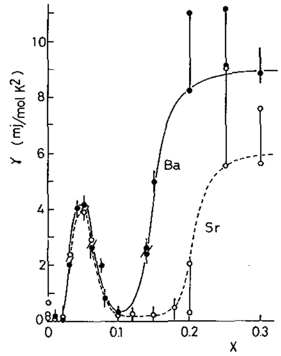

Fig. 3. – Concentration dependence of the coefficients of the T-linear term of heat capacity, γ, of $La_{2-x}Ba_xCuO_4$ (•) and $La_{2-x}Sr_xCuO_4$ (∘).

small humps of heat capacity near 6 K cause the rather scattered values for the concentrated Ba- and Sr-doping. (Superconducting transition temperature, T_c, show also a strong variation with Ba content, which we will discuss elsewhere.) The results of the vanishingly small γT values near $x = 0.1 \sim 0.2$ indicate that the contribution to the heat capacity from some parastic phases like $BaCuO_{2+x}$ is absent in the present La- system. No upturn behavior of heat capacity at low temperatures also supports the absence of impurity phases. We now understand that the poor reproducebility of the finite γ values reported so far is attributed to such strong variation of the γ values with x and to possible inhomogeneity of samples at the early studies.

At the present stage, several points should be noted for possible cause of the strong variation of γ. One possible interpretation of the very large γ for $x > 0.15$ (for Ba) and $x > 0.20$ (for Sr) is attributed to the structural inhomogeneity due to the transition from tetragonal structure at high temperature to orthorhombic one at low temperature near $x = 0.15$ for Ba and $x = 0.2$ for Sr [8].

Secondly, Shafer *et al.* [9] argued that T_c was described as a function of concentration of $[Cu - O]^{+p}$ charge, which increased initially with doping an decreased rapidly from $x \sim 0.15$ due to the increase of large oxygen deficiencies. It seems to be intertesting to clarify the relation between finite γ and the rapid derease of $[Cu - O]^{+p}$ charge for highly-doped samples.

Thirdly, a finite γ is connected to the existence of a pseudo-Fermi surface for spinons in term of the RVP picture [2]. However, the latter may exist only s-symmetry of the superconducting wave function. For the RVB wave function with d-symmetry which is found to be more stable at the mean field theory [10], there exists a sharp crossover-density $x_c \sim J\,/\,t$, through which the onset of superconductivity is characerized by different mecahnism [11]. The physical properties are markedly different in each side of this crossover. Recent theoretical study interprets that the zero γ in the superconducting state is possible within the RVB picture [12]. Finally, we have reported the absence of a linear term in Bi-Sr-Ca-Cu-O sample [13]. The T-linear term is not a general feature of the SC state of the new oxide superconductors.

Acknowledgments

The authors would like to thank Professor H. Fukuyama and Drs. P. Lederer and N. Wada for their helpful discussions. This work is supported in part by Grant-in-Aid for Scientific Research from the Minstry of Education, Science and Culture of Japan.

[1] Kumagai, K., Watanabe, I., Nakamura, Y., Aoki, H., Nakamichi, Y. and Nakajima, H., Proc. of the 4th Symp. on Magnetic and Magnetic Materials (Hsinchu, Taiwan) 1988, p. 287.

[2] Anderson, P. W., *Science* **235** (1987) 1196;
Baskaran, G., Zou, Z. and Anderson, P. W., *Solid State Commun.* **63** (1987) 973.

[2] Phillips, N. E. *et al.*, *Jpn J. Appl. Phys.* **26** (1987) 1115;
Reeves, M. E. *et al.*, *Phys. Rev. B* **35** (1987) 7207;
Dunlap, B. D. *et al.*, *Phys. Rev. B* **35** (1987) 7210;
Wenger, L. E. *et al.*, *Phys. Rev. B* **35** (1987) 7213;
Collecott, S. J. *et al.*, *Phys. Rev. B* **36** (1987) 5684.

[4] Ayache, C. *et al.*, *Solid State Commun.* **64** (1987) 247;
Ishikawa, M. *et al.*, *Physica B* **148** (1987) 332.

[5] Kumagai, K., Nakamichi, Y., Watanabe, I., Nakamura, Y., Nakajima, H., Wada, N. and Lederer, P., *Phys. Rev. Lett.* **60** (1988) 724.

[6] Kato, M., Maeno, Y. and Fujita, T., *Physica C* **152** (1988) 116.

[7] Kumagai, K., Watanabe, I., Aoki, H., Nakamura, Y., Kimura, T., Nakamichi, Y. and Nakajima, H., *Physica B* **148** (1987) 480.

[8] Fleming, R. M., Batlog, B., Cava, R. J. and Rietman, E. A., *Phys. Rev. B* **35** (1987) 7191.

[9] Shafer, M. W., Penny, T. and Olsen, B. L., *Phys. Rev.* **36** (1987) 4047.

[10] Kotliar, G., *Phys. Rev. B* **37** (1988) 3664.

[11] Suzumura, Y., Hasegawa, Y. and Fukuyama, H., *J. Phys. Soc. Jpn* **57** (1988) 401.

[12] Fukuyama, H., Hasegawa, Y. and Suzumura, Y., *Physica C* **152** (1988);
Suzumura, Y., Hasegawa, Y. and Fukuyama, submitted to *J. Phys. Soc. Jpn* **57** (1988) 2768.

[13] Kumagai, K. and Nakamura, Y., *Physica C* **152** (1988) 286.