Miniworkshop on
Non-Doped High Temperature Superconductors

- Nature of Superconductivity without Carrier Doping

July 9th, 2004, Tsukuba, Japan,
National Institute of Advanced Industrial Science and Technology (AIST). M604, 6th Fl., AIST Central 2

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Scope

Scientific progress has been often made by a sequence of creation and destruction. New concepts arise with the discarding of conventional views. Revolutionary changes in ideas may sometimes take place but the incubating period is usually long and things move only after a certain momentum has been gained. This is due to the fact that once principles are established as a mainstream, it is difficult to reevaluate them. However, to achieve a continuous progress careful attention must be directed to any indications that contradict with the principle are found.

High temperature superconductivity (HTSC) has been extensively studied since its discovery in the early 1980s, by an enormous number of researchers. The fundamental understanding among most of the researchers is that non-doped ground state of cuprates is a Mott insulator due to strongly correlated electrons in a nearly two-dimensional homogeneous conducting plane and the HTSC is observed only after doping carriers for which phonon is not taking a role. However, these fundamental aspects of HTSC are recently under criticism, i.e., the conductive plane is electronically inhomogeneous and the phonon contribution to the pairing mechanism cannot be neglected. Even the central issue, the nature of non-doped cuprates is under question. The recent observation that superconductivity can be found in nominally non-doped T' cuprates has raised a puzzling question on the fundamental concept of insulating state. Indeed, some of the Hubbard model calculations show that metallic electron state is realized depending on the strength of t/J, a key parameter of strongly correlated electron system. The objective of this workshop is to review those experimental results and theoretical calculations, and to propose a different view, i.e., the ground state without carrier doping may not always be insulating. All comments and questions are welcomed. The last part of the workshop will be dedicated to a round table discussion. We hope that the meeting will be fruitful in pointing us in the right direction.

July 9, 2004

Hiroyuki Oyanagi (AIST)

Michio Naito (TUAT)

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Program

13:30-13:40	**Opening remarks (Why now?)** H. Oyanagi, AIST

Chair: H. Eisaki, AIST

13:40-14:30	**Relation between local structure and transition temperature in high-Tc superconducting cuprates**, M. Naito, TUAT
14:30-15:00	**Key material issues in square-planar superconducting cuprates (T' and infinite-layer compounds),** A. Tsukada and S. Karimoto, NTT BRL
15:00-15:30	**Local structure of non-doped high Tc superconductors** H. Oyanagi, AIST
15:30-15:40	**Coffee Break**

Chair:S. Kashiwaya, AIST

15:40-16:10	**Fermi surfaces of cuprates superconductors: ARPES measurement and model HF calculation,** T. Mizokawa, Univ. of Tokyo
16:10-16:40	**Metal-insulator transition and relevance of models for high-Tc cuprates** T. Yanagisawa, AIST

Chair: T. Mizokawa, Univ. of Tokyo

16:40-17:10	**Two superconducting parameter domains of the 2D Hubbard model for high-Tc superconductors** K. Yamaji, AIST
17:10-17:30	**Open Discussion (Roundtable)**
17:30	**Closing remarks** M. Naito, TUAT
17:30-19:00	**Wine Party** (M604, 6th Fl., AIST Central 2)

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Relation between local structure and transition temperature in high-Tc superconducting cuprates

Michio Naito, Tokyo University of Agriculture and Technology

It has been believed for a long time that high-Tc superconductivity should be achieved by doping holes or electrons in the "perfect" CuO_2 planes. However, we have recently found a couple of examples that violate this scenario.

(1) Square-planar Nd_2CuO_4 and infinte-layer compounds do not show superconductivity even when they are hole-doped by excess oxygen.

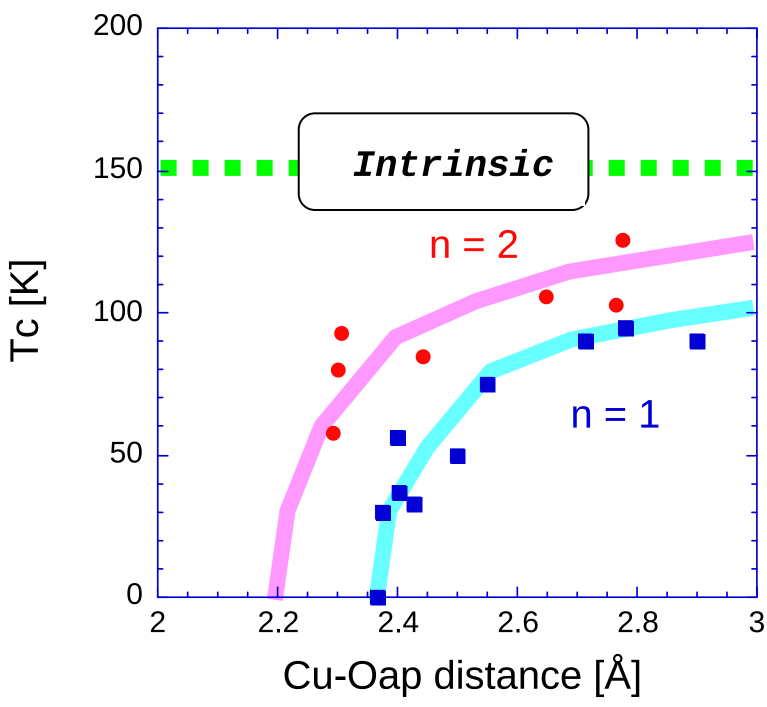
(2) Octahedral La_2CuO_4 does not show superconductivity even when it is electron-doped by Ce.

(3) Hole-doped $(La,Sr)_2CuO_4$ shows strain-dependent superconductivity. It shows Tc as high as 60 K with in-plane compressive and out-of-plane tensile strain but it hardly shows superconductivity with severe in-plane tensile and out-of-plane compressive strain.

These observations lead us to a new view for high-Tc superconductivity that is based on the local crystal structure. Our view is summarized as follows.

(1) The intrinsic Tc of the CuO_2 planes is ~150K.

(2) Apical oxygen is poison for high-Tc superconductivity. It reduces the intrinsic Tc roughly by 50K per one apical oxygen. Thereby pyramidal cuprates have Tc ~ 100 K, and octahedral cuprates have Tc ~ 50 K whereas (hole-doped) square-planar cuprates retains Tc ~ 150 K.



(3) The trend of Tc in pyramidal and

octahedral cuprates can be refined by taking account of D_{ap} (Cu-Oap distance). The empirical relation for T_c-vs-D_{ap} is shown in Fig. 1. The harmful role of apical oxygen in high-Tc superconductivity can be understood as creating pair-breaking Kondo scattering centers.

Key material issues in square-planar superconducting cuprates (T' and infinite-layer compounds)

A. Tsukada and S. Karimoto
NTT Basic Research Laboratories

Superconductivity in square-planar cuprates is very sensitive to impurity oxygen at the O(3) site (apical site), whose presence is detrimental to achieving superconductivity. As-grown T' cuprates ($Ln_{2-x}Ce_xCuO_4$) are not superconducting because they contain a small amount of impurity oxygen atoms at the apical site. Superconductivity appears only after heat treatment to remove apical oxygen. In the "well-known" bulk phase diagram of T' superconductors, the superconducting (SC) region is adjacent to the antiferromagnetic (AF) region, and the superconductivity suddenly appears at the SC-AF boundary with maximum T_c, suggesting competition between AF and SC orders. In the case of "optimally" doped $Pr_{1.85}Ce_{0.15}CuO_4$ or $Nd_{1.85}Ce_{0.15}CuO_4$, the AF correlations exist in as-grown non-superconducting samples, but they essentially disappear in reduced superconducting samples. This implies that more complete removal of apical oxygen would weaken the AF correlations and thereby expand the superconducting region. Actually, by improving the oxygen reduction process in the compound $Pr_{2-x}Ce_xCuO_4$, the superconducting region can be extended even with slightly increasing critical temperature down to a doping level of around x = 0.05. We believe that the apparent insulating behavior in the end-member T' compounds is not intrinsic but arises from the localization of carriers due to residual apical oxygen atoms. Therefore, the ρ-T behavior depends strongly on the concentration of residual apical oxygen atoms. With the same reduction recipe, the resistivity of T'-Ln_2CuO_4 becomes more metallic with larger Ln ions (Fig. 1). With the same compound, the ρ-T becomes more metallic with more complete removal of apical oxygen (Fig. 2).

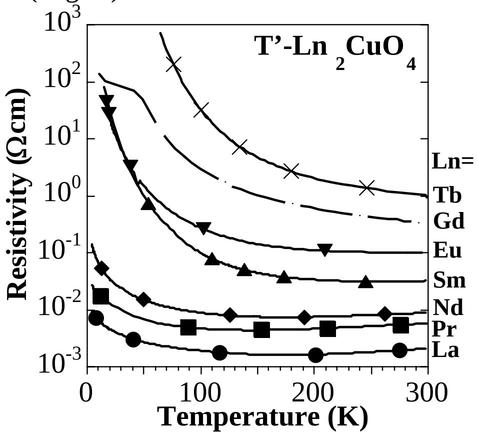


Fig. 1 ρ-T curves for T'-Ln_2CuO_4.

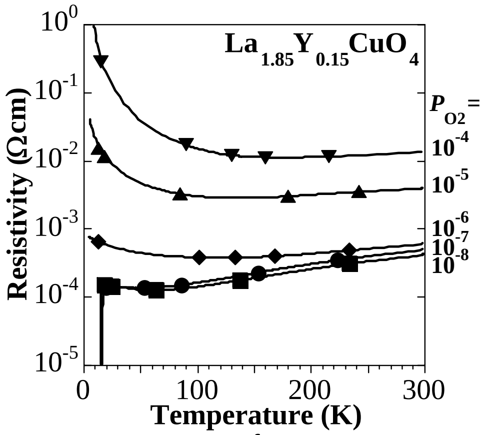


Fig. 2 ρ-T curves for $La_{1.85}Y_{0.15}CuO_4$ with different O_2 annealing atmosphere.

Local Structure of Non-Doped High Temperature Superconductors

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The recent report that epitaxially grown T' $(La,Y)_2CuO_4$ (LYCO) shows superconductivity (Tc~25 K) without intentional carrier doping[1] cast a challenge to our fundamental understanding of high temperature superconductivity (HTSC) that the ground state of non-doped cuprates is a Mott-insulator because of strong electron correlation, and HTSC is observed only after carrier doping[2]. In order to investigate the nature of unusual findings, we used x-ray absorption spectroscopy (XAS) to probe the short range order with a fast time scale (10^{-15} sec). The Cu K-edge x-ray absorption near-edge structure (XANES) and extended x-ray absorption fine structure (EXAFS) were measured for T' LYCO, T' $(La,Ce)_2CuO_4$ (LCCO) and T $(La,Sr)_2CuO_4$ (LSCO) as well as related bulk single crystals, Nd_2CuO_4 (NCO), $(Nd,Ce)_2CuO_4$ (NCCO) and $(Pr,Ce)_2CuO_4$, (PCCO).

Strongly anisotropic polarized Cu K-XANES spectra for 100 nm thick T' LYCO and LCCO showed signatures of T' structure, in sharp contrast to T LSCO. All XANES spectra for epitaxial thin films were in agreement with those of bulk counterparts, confirming the basic perovskite structure of $Ln_{2-x}RE_xCuO_4$ family. LSCO and LCCO revealed doping-dependent spectral variations. To our surprise, however, the E//*ab* and E//*c* polarized XANES of LYCO exhibited signatures of electron doping, *i.e.*, we found remarkable spectral similarity between LYCO and LCCO. It was found that Y doping induces the spectral shift and smearing of fine features found in LCCO, NCCO and PCCO, which suggests that a serious structural modification (disorder or heterogeneity) occurs on doping Y.

By Fourier transforming the E//*ab* EXAFS, the radial distribution functions (RDF) projected onto the basal plane were obtained. The results show that the Cu-O and Cu-Cu distances are elongated while RDF is broadened in LYCO and LCCO. Since doping rare earth element (Y) is believed to induce no charge carriers, we tentatively interpret the change of RDF as doping-induced local disorder. Decrease of instantaneous RDF for the Cu-O and Cu-Cu pairs could be due to nano-scale heterogeneity, *i.e.*, multi-domains having two different atomic spacings, *i.e.*, distorted domain around dopant atoms and unpurturbed domain. In summary, we observed remarkable similarity between non-doped and electron-doped T' cuprates in polarized XAS spectra on the Cu K-edge. A common feature, structural disorder or heterogeneity, seems to be important to understand the nature of metallic character of non-doped cuprates. In this talk, the results of polarized XAS experiments and the local structure will be discussed, in relation to the effect of doping on local structure.

1. M. Naito, S. Karimoto and A. Tsukada, Supercond. Sci. Technol. 15 (2002) 1663.
2. M. Imada, A. Fujimori and Y. Tokura, Rev. of Mod. Phys. 70 (1998) 1039.

Fermi Surfaces of Cuprates Superconductors: ARPES measurement and model HF calculation

T. Mizokawa

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Angle-resolved photoemission spectroscopy (ARPES) has been playing important roles in the study of high-T_c cuprates. In particular, doping dependence of Fermi surface and band dispersion has been revealed using the recently-developed high-energy-resolution ARPES [for example, A. Damascelli, Z. Hussain, Z.-X. Shen, Rev. Mod. Phys**.** 75, 473 (2003).]. In this talk, we will briefly review recent ARPES results on the electron/hole-doped systems and discuss the evolution of the Fermi surface based on the results of model Hartree-Fock calculation [D. Asakura and T. Mizokawa, Phys. Rev. B 68, 092508 (2003)]. In the slightly electron-doped regime, electron pockets emerge around (π, 0), which agrees with the ARPES study of $Nd_{2-x}Ce_xCuO_4$. In the slightly hole-doped regime, hole pockets emerge around (π/2, π/2) which might explain the photoemission result of $Ca_{2-x}Na_xCuO_2Cl_2$. The model calculation also predicts that the hole and electron pockets can coexist for the non-doped case. This would provide some insight on the recent studies of the non-doped systems.

Metal-insulator transition and relevance of models for high-Tc cuprates

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The microscopic origin of high-T_c superconductivity (SC) in cuprates has been studied intensively over the last fifteen years. We have not reached the solution for the mechanism of high-T_c SC and anomalous metallic behaviors, in which the correlation effects strongly dominate the physics. Although the parent compounds of all high-T_c cuprates have been considered as Mott insulators, recent experiments for rare-earth doped thin films have raised a question for this fixed idea [1]. Evidence of superconductivity in non-doped cuprates reported recently by NTT group leads us to reexamine the physics of parent compounds and lightly-doped materials. The physics of non-doped parent compounds is closely related to a question "which models are relevant for investigation of high-T_c cuprates ?". We discuss this problem comparing several models such as the t-J model and Hubbard model.

The results for non-doped cuprates reported in [1] has opened a way for the Hubbard model to be a basic model for high-T_c SC. The Hubbard model is a fundamental model to describe the metal-insulator transition for the non-doped system. We have a possibility to control the metal-insulator transition by tuning the strength of the Coulomb repulsion U and the transfer integral t' to change the Fermi surface shape. For the doped case, the overall structure of phase diagram is understandable based on calculations using the quantum variational Monte Carlo method [2-4]. A possibility of superconductivity within the Hubbard model, however, still remains the problem. This issue is also discussed using a quantum Monte Carlo method.

[1] A. Tsukada et al., cond-mat/0311380, cond-mat/0401120.
[2] K. Yamaji et al., Physica C304, 225 (1998); Physica C392-396, 229 (2003).
[3] T. Yanagisawa et al., Phys. Rev. B64, 184509 (2001); B67, 132408 (2003).
[4] M. Miyazaki et al., J. Phys. Soc. Japan 73 (2004).

Two Superconducting Parameter Domains of the 2D Hubbard Model for High-T_c Superconductors

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We have been studying the two-dimensional (2D) Hubbard model to understand the microscopic origin of high-T_c superconductivity (SC) in cuprates. We looked for the lowest-energy state by computing the condensation energies E_{cond} of *d*-wave SC and SDW states by means of the variational Monte Carlo method. Based on recent analyses of observed spin-wave energies in La_2CuO_4 we employed moderate on-site Coulomb energies U ~ 6 (in units of nearest-neighbor transfer energy t) [1]. Taking account of the second-neighbor transfer energy t' we found that, when hole-doping is optimal, SC dominates SDW in the region of $-0.1 < t' < 0$ [2]. The obtained SC E_{cond} was in reasonable agreement with experimental values. In view of the estimated value of t' ~ –0.15 [3] the results are considered to explain the SC in La214. In the region of $-0.5 < t' < -0.1$ the SDW E_{cond} is overwhelmingly larger than the SC E_{cond}, making SC impossible to survive in this region. However, some authors estimated the value of t' at ~ –0.3 together with third–neighbor transfer energy t'' ~ 0.2 for YBCO and Bi2212 [3]. Recently we noticed that the presence of t'' makes the situation different. In this case a combined parameter $t'-2t''$ controls the nesting of the Fermi surface, consequently the strength of SDW, instead of t' in the case of the simple model without t''. Since $t'-2t'' < -0.5$ is satisfied, SDW is expected to vanish or vanishingly weaken and so SC should be stable for optimal doping. Our computation confirmed this inference; SDW disappeared and SC was found to be remarkably enhanced in the parameter domain around t' ~ –0.3 and t'' ~ 0.2, presumably due to the large state density around (π, π). This result makes the SC in YBCO and Bi2212 understandable in the present scheme. For certain sets of t' and t'' in this parameter domain SC was found to win against SDW even with no doping. This is a plausible reason enabling superconductivity in non-doped cuprates reported in [4].

[1] R. Coldea et al., Phys. Rev. Lett. 86, 5377 (2001); N. M. Peres et al. Phys. Rev. B 65, 132404 (2002); P. Sengupta et al., Phys. Rev. B 66, 144420 (2002).
[2] K. Yamaji et al., Physica C 392-396, 229 (2003); Physica C in press.
[3] T. Tanamoto et al., J. Phys. Soc. Jpn. 61, 1886 (1992); T. Tohyama et al., Supercond. Sci. Technol. 13, R17 (2000).
[4] A. Tsukada et al., cond-mat/0311380 (2003); H. Yamamoto et al., Physica C in press [cond-mat/040119]; A. Tsukada et al., Physica C in press [cond-mat/0401120].
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