# Solid State Chemistry - Quantum Spin Fluids -



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Quantum spin oxide system such as high- $T_c$  superconducting cuprates, La<sub>2-x</sub>(Ba,Sr)<sub>x</sub>CuO<sub>4</sub>, Nd<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4</sub> and Pr<sub>1-x</sub> LaCe<sub>x</sub>CuO<sub>4</sub> are synthesized in the form of single crystals using traveling-solvent-floating-zone method. Detailed equilibrium phase diagram of Bi cuprate systems is investigated. Main subjects and techniques are: mechanism of high- $T_c$  superconductivity: origin of quantum phase separation in strongly correlated electron systems: spin excitations in quantum spin systems: interplay between spin and charge flow in doped spin systems: neutron scattering by using triple-axis as well as time-of flight-techniques.

# **Research Activities (Year 2001)**

#### Presentations

Magnetic phase diagrams of electron-doped high- $T_c$  cuprates studied by  $\mu$ SR measurements, T. Kubo, M. Fujita, T. Uefuji, K. Yamada, I. Watanabe, K. Nagamine, Autumn Meeting, Phys. Soc. Jpn., 18 Sep., ISS 2001, 26 Sep.

Preparation of electron-doped superconductor by electrochemical reduction, K. Kawashima, M. Fujita, T. Kubo, K. Yamada, Autumn Meeting, Phys. Soc. Jpn., 18 Sep., ISS 2001, 26 Sep.

Structural effect on the stability of magnetic order in La<sub>1.875</sub>Ba<sub>0.125-x</sub>Sr<sub>x</sub>CuO<sub>4</sub>, H. Goka, T. Kubo, T. Uefuji, M. Fujita, K. Yamada, M. Matsuda, I. Watanabe, K. Nagamine, Autumn Meeting, Phys. Soc. Jpn., 18 Sep., ISS 2001, 26 Sep.

Electron-doping effect on magnetic order and superconductivity in Nd<sub>2x</sub>Ce<sub>x</sub>CuO<sub>4</sub>, T. Uefuji, K. Kurahashi, M. Fujita, M. Matsuda, K. Yamada, Autumn Meeting, Phys. Soc. Jpn., 17 Sep., ISS 2001, 26 Sep.

#### Grants

Yamada K, Study of quantum phase separation in the transition metal oxides, Grant-in-Aid for Scientific Research on Priority Areas (Novel Quantum Phenomena in Transition Metal Oxides), 1 April 2000 - 31 March 2003.

Yamada K, Study of magnetic correlation and cooperative/competitive phenomena of electronic conductivity in the localized/itinerant electron systems, Grant-in-Aid for Scientific Research (A), 1 April 2000 - 31 March 2002.

Fujita M, Study of High- $T_c$  superconductivity mechanism in the electron-doped cuprates, Grant-in-Aid for Encouragement of Yong Scientists, 1 April 2001 - 31 March 2003.

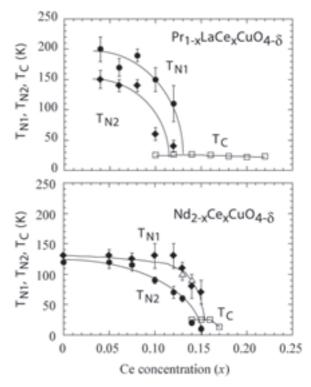
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## Magnetic phase diagrams of NCCO and PLCCO investigated by means of µSR measurements

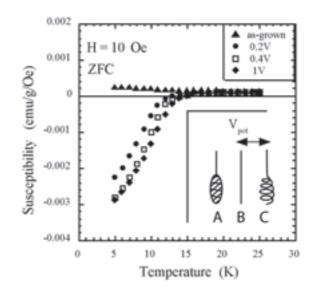
Magnetic phase diagrams of electron-doped high-temperature superconducting cuprates Nd<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4-δ</sub> (NCCO) and Pr1-xLaCexCuO4-8 (PLCCO) have been studied by means of muon spin relaxation and rotation ( $\mu$ SR) measurements. Two characteristic temperatures  $T_{N1}$  and  $T_{\rm N2}$  are defined in the zero-field  $\mu SR$  (ZF- $\mu SR$ ) measurements for both NCCO and PLCCO. Below  $T=T_{N1}$ , which approximately corresponds to the previously reported Néel temperature, an exponential type muon spin relaxation firstly appears in the time spectra and upon cooling below around  $T_{N2}$  another faster component of relaxation or muon spin rotation is observed similarly. Although critical concentration for the superconductivity is different between the two systems, antiferromagnetic (AF) phase commonly contacts with the superconducting phase. In contrast to the hole-doped superconductivity the electron-doped superconductivity is more drastically terminated by the AF order.

### Preparation of electron-doped superconductor by electrochemical reduction

Electrochemical reduction processes were performed using the opposite chemical reactions to the electrochemical oxidation. As shown in the right figure, we succeeded in preparing superconducting Nd<sub>1.85</sub>Ce<sub>0.15</sub>CuO<sub>4</sub> samples from the insulating as-grown samples using electrochemical reduction, though the obtained  $T_c \sim 15$ K is lower than that of the heat-treated sample,  $T_c \sim 24$ K. In this figure, we show the dependence on  $V_{\text{pot}}$  of magnetic susceptibilities for samples treated with different  $V_{pot}$  values,  $V_{\text{pot}} = V_{\text{count}}$  -  $V_{\text{ref}}$  between the counter and reference electrodes in the range of  $0 \sim 1V$  as shown in the inset. We, furthermore, performed the electrochemical reduction under various conditions as functions of  $V_{pot}$  and treating time. However, the maximal diamagnetism at 5K and  $T_{\rm c}$  were nearly the same. One of the reasons for such small diamagnetic signal and low  $T_{\rm c}$  compared to heattreated samples may be due to a small mobility of oxygen atoms inside the sample, which causes the inhomogeneous distribution of oxygen atoms.



Magnetic phase diagrams of PLCCO and NCCO.



Magnetic susceptibilities of  $Nd_{1.85}Ce_{0.15}CuO_4$  samples reduced at various  $V_{pot}$  for 1 day. The inset shows a schematic drawing of the configuration of three electrodes. A, B and C denote a working electrode, a reference electrode and a counter electrode, respectively.