

# Re-appearance of antiferromagnetic ordering with Zn and Ni substitution in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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## Abstract

The effects of nonmagnetic Zn and magnetic Ni substitution for Cu site on magnetism are studied by measurements of uniform magnetic susceptibility for lightly doped  $\text{La}_{2-x}\text{Sr}_x\text{Cu}_{1-z}\text{M}_z\text{O}_4$  (M=Zn or Ni) polycrystalline samples. For the parent  $x=0$ , Zn doping suppresses the Néel temperature  $T_N$  whereas Ni doping hardly changes  $T_N$  up to  $z=0.3$ . For the lightly doped samples with  $T_N \sim 0$ , the Ni doping recovers  $T_N$ . For the superconducting samples, the Ni doping induces the superconductivity-to-antiferromagnetic transition (or crossover). All the heavily Ni doped samples indicate a spin glass behavior at  $\sim 15$  K.

*Key words:* impurity effect; magnetic susceptibility;  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ; superconductor-to-antiferromagnet crossover

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Although nonmagnetic impurity Zn substitution effect has been extensively studied for high- $T_c$  cuprate superconductors and the parent Mott insulators, magnetic impurity Ni substitution effect has not been extensively studied relatively. Particularly, to our knowledge, there are a few studies for Ni doping effect in semiconducting regime ([1–4]). In this paper, we report a systematic study of Ni substitution effect on the polycrystalline samples of  $\text{La}_{2-x}\text{Sr}_x\text{Cu}_{1-z}\text{M}_z\text{O}_4$  in the parent antiferromagnet, the lightly doped insulators without long range order, and the relatively low- $T_c$  superconductors, through measurement of uniform magnetic susceptibility  $\chi$ . The polycrystalline samples were synthesized by a solid state reaction method. For comparison, we synthesized also  $\text{La}_{2-x}\text{Sr}_x\text{Cu}_{1-z}\text{Zn}_z\text{O}_4$  [5]. Here, we emphasize an importance of careful annealing process at 650 °C for 48 hours under Ar gas atmosphere. The uniform magnetic susceptibility was mea-

sured by a SQUID magnetometer. The Néel temperature  $T_N$  is determined by the maximum behavior, or the onset temperature of hysteresis of the magnetic susceptibility between zero field cooling (ZFC) and field cooling (FC). The spin glass temperature  $T_{SG}$  is defined by the low temperature sharp peak in the further hysteresis [6]. For non-superconducting samples, a magnetic field of  $100 \sim 1.0 \times 10^4$  Oe was applied, whereas for superconducting samples, a field of  $\sim 100$  Oe was applied.

Figure 1 shows Ni doping effect on the  $T$  dependence of magnetic susceptibility. We found the followings:

1) Up to  $z=0.3$  for pure  $\text{La}_2\text{CuO}_4$ , Ni doping does not destroy the Néel ordering. Such a robust  $T_N$  to Ni doping is in contrast to a fragile  $T_N$  to Zn doping [7,3]. In Fig. 2, for comparison,  $T_N$  versus Ni or Zn content  $z$  is shown.

2) With further Ni doping  $z > 0.3$ , the spin glass ordering appears at  $T_{SG} \sim 15$  K, probably due to Ni spin freezing. Hereafter, we call this Ni freezing temperature.

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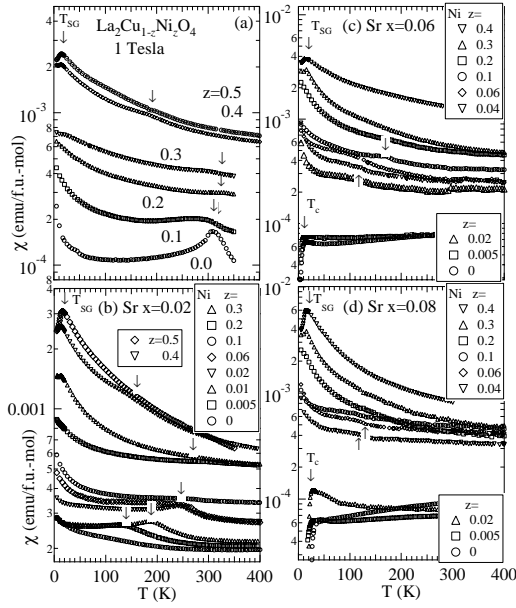


Fig. 1. Ni-doping effect on the ZFC *dc* magnetic susceptibility of  $\text{La}_{2-x}\text{Sr}_x\text{Cu}_{1-z}\text{M}_z\text{O}_4$ ; parent insulator  $x=0$  (a), lightly doped non-superconducting  $x=0.02$  without long range order at  $z=0$  (b), lightly doped  $x=0.06$  with relatively low  $T_c$  at  $z=0$  (c), and  $x=0.08$  at  $z=0$  (d). The arrows without character indicate  $T_N$ 's, the other arrows with character are  $T_c$ 's, or  $T_{SG}$ 's. For simplicity, we do not attach all the arrows.

3) The Néel ordering, which is suppressed down to  $T_N < 4.2$  K by Sr doping  $x=0.02$ , recovers more rapidly and largely with Ni doping, than with Zn doping [5].

4) The superconductivity for Sr  $x=0.06$  or  $0.08$  is easily suppressed by Ni doping. The Ni doping induces the superconductor-to-antiferromagnet transition (crossover) at  $z=0.02\sim 0.04$ . Further Ni doping for  $z > 0.3$  or  $> 0.2$  induces the spin glass state with  $T_{SG} \sim 15$  K.

5) The Ni freezing temperature with heavily Ni doping does not seem to depend on Sr doping level.

6) The Ni freezing temperature  $T_{SG} \sim 15$  K is about two times larger than the Ni-free, spin glass temperature  $T_g \sim 7$  K at Sr  $x=0.04$  [6].

In Fig. 3, we summarize the magnetic phase diagram versus Ni content  $z$  at various Sr doping, which can be drawn from the preset study in Fig. 1. The magnetic impurity Ni doping yields rich phases through the order-to-disorder transition (or crossover) in antiferromagnetic correlation, the spin glass transition on Ni spin freezing, and the superconductor-to-insulator transition (or crossover). In conclusion, we demonstrate that Ni doping causes the above novel effects on the strongly correlated electron system  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

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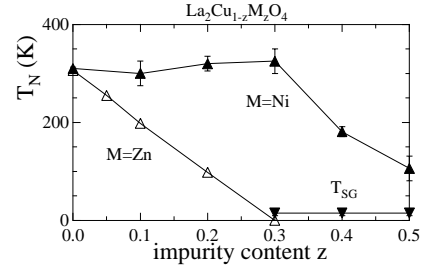


Fig. 2.  $\text{La}_2\text{Cu}_{1-z}\text{M}_z\text{O}_4$ :  $T_N$  versus M=Ni or Zn content  $z$ . The result is qualitatively consistent with Refs. [7,3].

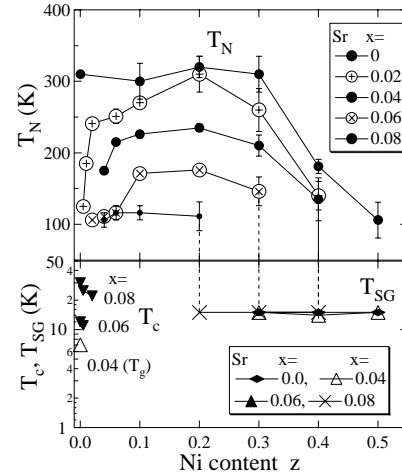


Fig. 3. Magnetic phase diagram of  $\text{La}_{2-x}\text{Sr}_x\text{Cu}_{1-z}\text{M}_z\text{O}_4$ ;  $T_N$ ,  $T_{SG}$ ,  $T_g$ , and  $T_c$  versus Ni content  $z$  at Sr doping. The solid and the dashed lines are guide for the eye.

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