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

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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 La$_{2-x}$Sr$_x$Cu$_{1-z}$M$_z$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 $T_N$~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.

74.72.Dn, 75.40.Cx, 75.30.Hx

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 La$_{2-x}$Sr$_x$Cu$_{1-z}$M$_z$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 La$_{2-x}$Sr$_x$Cu$_{1-z}$Zn$_z$O$_4$ [5]. Here, we emphasize an importance of careful annealing process at 650 $^\circ$C for 48 hours under Ar gas atmosphere. The uniform magnetic susceptibility was measured 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 100~1.0×10$^4$ 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 La$_2$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.

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 $z=0.02$~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 La$_{2-x}$Sr$_x$CuO$_4$.

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FIG. 1. Ni-doping effect on the ZFC dc magnetic susceptibility of La$_{2-x}$Sr$_x$Cu$_{1-z}$M$_z$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.

FIG. 2. La$_{2}Cu_{1-z}M_zO_4$; $T_N$, $T_{SG}$, $T_g$, and $T_c$ versus Ni content $z$ at Sr doping. The solid and the dashed lines are guide for the eye.

FIG. 3. Magnetic phase diagram of La$_{2-x}$Sr$_x$Cu$_{1-z}$M$_z$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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