Novel in-gap state in Zn-doped $La_{1.85}Sr_{0.15}CuO_4$

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It is a received wisdom that the antiferromagnetism (AF) on a hole-doped CuO₂ plane in a lamellar copper oxide is relevant to the high T_c superconductivity. Our aim is to understand the spin state of doped CuO_2 plane, particularly in the superconducting state. However, there exists no AF long-range order (LRO), and the low-energy spin excitations, which might play an important role in the high T_c superconductivity, disappear due to a so-called *spin qap* at low temperatures. The best way to observe the low-energy spin response is to suppress the superconductivity by giving some perturbations to the spin system without changing the other properties. Zn doping is extremely suitable for the purpose: A small amount of Zn^{2+} ions substituting for Cu^{2+} ions strongly suppress the superconductivity without doping holes nor giving much structural disturbance. NMR and neutron studies have revealed that doped Zn impurities induce staggered magnetic moments on the surrounding Cu sites, indicating that Zndoping enhances AF correlations on a CuO_2 plane. [1, 2] If we can tune T_c by precisely controlling the Zn concentration in single crystals large enough for inelastic neutron scattering, it would provide a chance to study in detail the microscopic nature of spin correlations and their contribution to the high T_c paring mechanism.

We have carried out a comprehensive neutron scattering study of the AF spin correlations in Zn-doped $La_{1.85}Sr_{0.15}CuO_4$ single crystals to elucidate how the spin-gap state is broken and how the static AF correlations are induced by Zn-doping. To obtain quantitative information about the Zn-doping dependence of spin excitation spectra, it is essential to control the Zn-doping rate accurately. Furthermore, large and spatially homogeneous crystals are required. We have overcome such difficulties by combining the traveling-solvent-floatingzone (TSFZ) method and a quantitative analysis of elements using the inductively-coupled plasma method. Our systematic studies with changing the Zn-doping rate under *unified* experimental conditions revealed a novel low-energy spin excitation which is induced within the spin gap state by doped Zn impurities.

Figure 1 shows the energy spectra of the dynamical spin susceptibility $\chi''(\omega)$ around 10 K. All the data are corrected so as to be directly compared. It is remarkable that $\chi''(\omega)$ for all the samples have a maximum and almost identical intensity around $\omega = 8$ meV, while $\chi''(\omega)$ below 8 meV develops with increasing doped Zn. These systematic changes cannot be explained by either the broadening of the gap structure or the reduction of the gap-energy because both the cases should be associated with the variation of $\chi''(\omega)$ near the gap energy. Therefore, these energy spectra indicate that Zn-doping induces an *additional* spin excitation in the spin gap below $\omega \sim 8$ meV and that, with further doping, the novel spin excitation is enhanced and shifts to lower energies, i.e., becomes more static. Indeed, the elastic magnetic signals were observed for y = 0.017 below ~ 20 K at the



Figure 1: Energy dependence of the q-integrated $\chi''(\omega)$ for (a) y = 0.004, (b) y = 0.008 and (c) y = 0.017 at 10 K. Open diamonds denote the data for y = 0.0. Solid lines are fits with a phenomelogical dynamical spin susceptibility.

same incommensurate positions as that of the inelastic scattering, while no signal was detected in y = 0.008 down to 1.5 K. The in-plane spin correlation length for y = 0.017 is estimated at ~ 80 Å.

A mean distance between Zn ions (R_{Zn-Zn}) shortens from ~ 60 Å in y = 0.004 to ~ 42 Å in y = 0.008. We thus speculate that the induced local magnetic moments around a doped Zn ion start correlating with those around other Zn ions for y = 0.008, and that the correlations among the moments around different Zn ions become coherent, which gives rise to the novel in-gap spin state at particular q positions. The in-plane *static* spin correlation lengths for y = 0.014 and 0.017 exceed 80 Å, which is much longer than their R_{Zn-Zn} values, 32 and 29 Å. This fact indicates that the static spin correlations originate not from the independent local magnetisms around Zn impurities but from the long-range AF coherence among the induced moments around different Zn ions. Thus we conclude that the in-gap state continuously connects with an AF ground state with increasing Zn, i.e., decreasing R_{Zn-Zn} , indicating the importance of underlying AF ground state which is locally substituted for the superconducting state with help of small perturbations.

References

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