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# Magnetic properties of the $S$=1/2 quasi-one-dimensional antiferromagnet: $Sr_{14-x}Y_xCu_{24}O_{41}$



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Magnetic susceptibility and inelastic neutron scattering measurements have been performed on the $S$=1/2 quasi-one-dimensional system $Sr_{14-x}Y_xCu_{24}O_{41}$, which has both simple chains and two-leg ladder chains of copper ions. The experimental results show that the simple chain in $Sr_{14}Cu_{24}O_{41}$ has a spin gap in the excitation spectrum, which originates from a dimerized state. We have also studied the effect of $Y^{3+}$ substitution for $Sr^{2+}$ site on the dimerized state. It was found that the yttrium substitution suppresses the gap energy drastically.

The structure of $Sr_{14}Cu_{24}O_{41}$ [1, 2] consists of two unique subcells. One is chains of copper ions which are coupled by the nearly 90° Cu-O-Cu bonds. The other is two-leg ladders of copper ions, which are coupled by the nearly 180° Cu-O-Cu bonds along the $a$ and $c$ axes. Each ladder is coupled by the nearly 90° Cu-O-Cu bonds. The interaction between the ladders is considered to be much weaker than that within the ladder. Each chain and Sr ions form layered structure in the $ac$ plane and stack alternately along the $b$ axis.

The yttrium substitution for strontium site affects the crystal structure and the electronic states. The yttrium doping decreases the lattice constants. Since the structure is layered, the substitution directly affects the distance between the layers along the $b$ axis with little effect on $ac$ plane parameters which are determined by the copper-oxygen connectivity. It is noted that the valence state of copper ions in the stoichiometric $Sr_{14}Cu_{24}O_{41}$ is +2.25, suggesting the existence of holes at the oxygen sites. The hole carriers are considered to be localized because the compound is highly insulating [1, 3]. The trivalent yttrium substitution for divalent strontium is expected to decrease hole carriers. The lanthanide substitution also causes small distortion to both the chain and the ladder.

The results of magnetic susceptibility measurements on $Sr_{14}Cu_{24}O_{41}$ have shown a broad peak below 100 K [3-6]. The nearly 180° Cu-O bondlength (1.90 Å perpendicular to the ladder and 1.97 Å along the ladder) of the two-leg ladders in $Sr_{14}Cu_{24}O_{41}$ is close to the corresponding value in $SrCu_2O_3$ (1.93 Å perpendicular to the ladder and 1.97 Å along the ladder). Then it is natural to assume that the gap energy of the ladder in $Sr_{14}Cu_{24}O_{41}$ is close to that of $SrCu_2O_3$ (420 K from susceptibility measurements and 680 K from NMR measurements [7]). This means that the ladder in $Sr_{14}Cu_{24}O_{41}$ has a singlet ground state with a fairly large gap and that the magnetic susceptibility from the ladder is very small below room temperature. Therefore, the results of the susceptibility measurements suggest that there is a spin gap originating from the chain in this compound.

The powder samples of $Sr_{14-x}Y_xCu_{24}O_{41}$ ($x$ = 0, 1 and 3) were prepared by firing stoichiometric ratio of $SrCO_3$ (99.99%), $Y_2O_3$ (99.99%) and CuO (99.99%) at 980 °C for 30 h in air with intermittent regrinding. Powder x-ray diffraction experiments were performed to check that the reaction was complete. The magnetic susceptibility was measured by using a SQUID magnetometer (Quantum Design MPMS2). The inelastic neutron scattering experiments were carried out on the H7 and H8 triple-axis spectrometers at the High Flux Beam Reactor at the Brookhaven National Laboratory (BNL). We used similar volume of the powder samples to perform in-

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elastic neutron measurements. The scattering intensities from the three different powder samples are similar within an error of 10%.

Figure 1 shows the magnetic susceptibility in the powder samples of $Sr_{14-x}Y_xCu_{24}O_{41}$ ($x$ = 0, 1 and 3). The solid line shows the susceptibility of the $x$=0 sample after a low temperature Curie tail was subtracted. This shows a spin gap (~100 K) in the excitation spectrum originating from the simple chains as descibed above. The Curie tail increases as the yttrium is substituted for strontium site. In the $x$=1 sample a small anomaly is seen at ~30 K, below which the susceptibility rapidly increases. In the $x$=3 sample the susceptibility monotonically decreases with increasing temperature. The results suggest that the spin gap disappears between $x$=1 and 3.

We show in the inset of Fig. 1 inelastic neutron scattering spectra at $Q$=1.12 $\AA^{-1}$ measured at $T$=10 K in the powder samples of $Sr_{14-x}Y_xCu_{24}O_{41}$ ($x$=0, 1 and 3). A flat-topped inelastic peak is observed between 10 and 14 meV in the $x$=0 sample [8]. The intensity decreases with increasing temperature. As the yttrium is substituted for strontium site, the gap energies are suppressed. The $x$=1 sample shows a

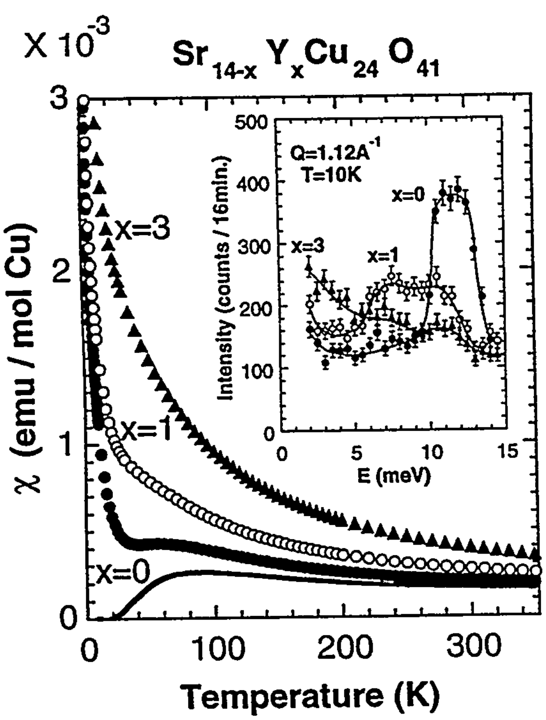


Fig. 1. Temperature dependence of magnetic susceptibility in the powder samples of $Sr_{14-x}Y_xCu_{24}O_{41}$ ($x$ = 0, 1 and 3). The inset shows the inelastic neutron scattering spectra at $Q$=1.12 $\AA^{-1}$ measured at $T$=10 K (from ref. 8).

broader flat-topped peak between 6 and 13 meV and the peak intensity becomes about half of the $x$=0 sample, although the integrated intensity is almost the same as the $x$=0 sample. The $x$=3 sample does not show any peaks but a tail which decreases with increasing energy.

We now discuss the substitution effect on the spin gap. A possible origin of this spin gap has been discussed by Matsuda and Katsumata as arising from a dimerized state [6]. If the small decrease of the lattice constants with the substitution affects the dimerized states, the superexchange interaction between $Cu^{2+}$ spins becomes stronger and so the gap energy increase. This suggests that the decrease of the holes mainly affect the dimerized states. The number of holes in the chain is 60% of that of Cu ions in the chain of $Sr_{14}Cu_{24}O_{41}$, 50% in $Sr_{13}Y_1Cu_{24}O_{41}$ and 30% in $Sr_{11}Y_3Cu_{24}O_{41}$. This drastic change of hole number will change the electronic states of Cu and oxygen dramatically. The decrease of hole number probably makes the dimerized state unstable. Then the chains in $Sr_{14-x}Y_xCu_{24}O_{41}$ behave like an $S$=1/2 1D Heisenberg antiferromagnet which has no gap in the excitation spectra.

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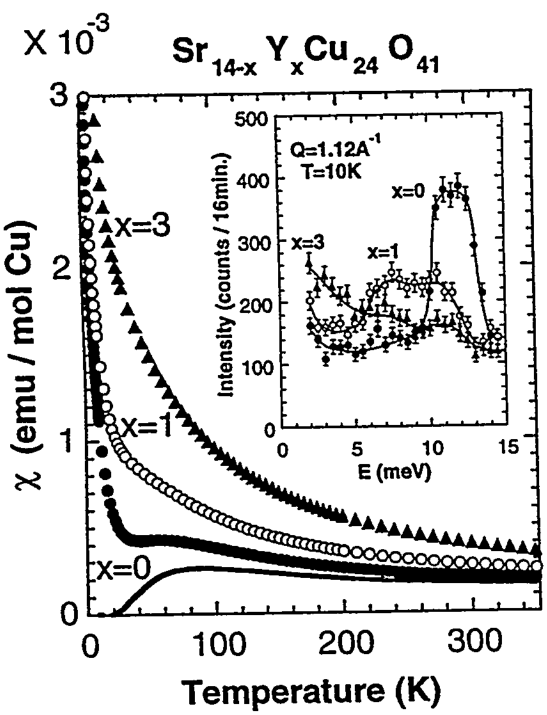


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