Magnetic excitations and structural change in
the $S=\frac{1}{2}$ quasi-one-dimensional magnet
$\text{Sr}_{14-x}\text{Y}_x\text{Cu}_{24}\text{O}_{41}$ ($0 \leq x \leq 1$)

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Neutron scattering measurements have been performed on the $S=\frac{1}{2}$ quasi-one-dimensional system $\text{Sr}_{14-x}\text{Y}_x\text{Cu}_{24}\text{O}_{41}$, which has both simple chains and two-leg ladders of copper ions. We observed that when a small amount of yttrium is substituted for strontium, which is expected to reduce the number of holes, the dimerized state and the structure in the chain are changed drastically. The inelastic peaks originating from the dimerized state of the chain becomes broader in energy but not in momentum space. This implies that the dimerized state becomes unstable but the spin correlations are unchanged with yttrium substitution. Furthermore, it was observed that nuclear Bragg peak intensities originating from the chain show strong temperature and $x$ dependence, which suggests that the chains slide along the $c$ axis as temperature and $x$ are varied.

75.25.+z, 75.10.Jm, 75.40.Gb

I. INTRODUCTION

As a byproduct of the high temperature superconducting copper oxides, many interesting quasi-one-dimensional copper oxides have been discovered or rediscovered recently. $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ is one of them and consists of two kinds of unique building blocks as shown in Fig. 1a. One is simple chains of copper ions which are coupled by the nearly $90^\circ$ Cu-O-Cu bonds. The other is two-leg ladders of copper ions, which are coupled by the nearly $180^\circ$ Cu-O-Cu bonds along the $a$ and $c$ axes. This compound has been extensively studied since both of the building blocks show interesting ground states.

The ground state of a two-leg spin ladder system is a singlet state as observed in $\text{Sr}\text{Cu}_2\text{O}_3$. It was also shown that the ladder in the related compound, $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ has a singlet ground state with a fairly large gap. The property of the energy gap in spin ladders is interesting from the view point of quantum phenomena in a low-dimensional (between 1 and 2) Heisenberg antiferromagnet. It was theoretically predicted that the spin $\frac{1}{2}$ Heisenberg ladder with even numbers of legs has an excitation gap and that the excitation is gapless for the spin ladder with odd numbers of legs. The two-leg ladder system has also attracted many researchers since superconductivity is expected in the carrier doped spin ladder system. Recently, Uehara et al. found that $\text{Sr}_{10.4}\text{Ca}_{13.6}\text{Cu}_{24}\text{O}_{41}$ shows superconductivity below 12 K under a high pressure of 3 GPa. The superconductivity in the ladder system is considered to be crucial to understand the mechanism of the high temperature superconductivity.

As Matsuda et al. showed, the simple chain in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ also has an interesting singlet ground state originating from a dimerization. Surprisingly, the dimers are formed between spins which are separated by 2 and 4 times the distance between the nearest-neighbor (n.n.) copper ions in the chain. This is probably related to localized holes in the chain which are expected to make the interaction between copper spins longer-ranged. The trivalent yttrium substitution for divalent strontium is expected to decrease hole carriers. The dimerized state in the chain depends critically on the number of holes ($N_h$). The magnetic inelastic peaks become broader in energy with yttrium substitution. Furthermore, $\text{La}_{6}\text{Ca}_{8}\text{Cu}_{24}\text{O}_{41}$ in which $N_h=0$ shows a long-range magnetic order with ferromagnetic correlations within the chain.

We have performed neutron scattering experiments to study the effects of yttrium substitution on the magnetic and structural properties of the chains in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$. We previously reported the results of neutron scattering...
experiments using polycrystalline samples of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0, 1, \text{and } 3$) in order to make a detailed study of the magnetic excitation in a wide range of energy ($\omega$) - momentum ($Q$) space and the crystallographic structure, we performed new experiments on high quality single crystals. Furthermore, we concentrated on the samples with low concentration of yttrium to study systematically how the dimerized state is changed since the dimerized state is destroyed with only a small amount of yttrium substitution. It was observed that when yttrium is lightly substituted for strontium, strong and sharp magnetic inelastic peaks which originate from the dimerized state in the chain become broader. With further yttrium substitution, the inelastic peaks become much broader and the excitation energy is decreased. The interesting point is that the inelastic peaks become broader only in energy but not in momentum space. This means that the dimerized state becomes unstable but the spin correlations are unchanged with yttrium substitution. It was also observed that nuclear Bragg intensities originating from the chain show strong temperature and yttrium concentration dependence. One possible explanation for this would be that the chains shift along the $c$ axis with temperature and yttrium substitution.

The format of this paper is as follows: Experimental details are described in Sec. II. The magnetic and structural studies are presented in Sec. III and IV, respectively. In Sec. V the experimental results are discussed.

II. EXPERIMENTAL DETAILS

The single crystals of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0, 0.10, 0.25,$ and $1.0$) were grown using a traveling solvent floating zone (TSFZ) method at 3 bars oxygen atmosphere. The effective mosaic of the single crystals is less than $0.4^\circ$ with the spectrometer conditions as is described below. It is expected that yttrium is distributed homogeneously in the sample since the lattice constant $b$ is systematically decreased and the linewidth of the nuclear Bragg peaks does not change with yttrium substitution. The lattice constant $b$ is $13.36$ Å and $13.08$ Å at $10$ K for the $x=0$ and $x=1$ samples, respectively.

The neutron scattering experiments were carried out on the HB3 triple-axis spectrometer at the High Flux Isotope Reactor at Oak Ridge National Laboratory and the H8 triple-axis spectrometer at the High Flux Beam Reactor at Brookhaven National Laboratory. The horizontal collimator sequence was open-$40^\circ$-$S$-$60^\circ$-$120^\circ$ for the experiments on the HB3 and $40^\circ$-$40^\circ$-$S$-$80^\circ$-$80^\circ$ on the H8. The neutron measurements of Sr$_{14}$Cu$_{24}$O$_{41}$, Sr$_{13.5}$Y$_{0.1}$Cu$_{24}$O$_{41}$, and Sr$_{13.75}$Y$_{0.25}$Cu$_{24}$O$_{41}$ were performed on HB3 and Sr$_{13}$Y$_1$Cu$_{24}$O$_{41}$ on H8. The final neutron energy was fixed at $E_f=14.7$ meV. Pyrolytic graphite crystals were used as monochromator and analyzer; contamination from higher-order beam was effectively eliminated using a pyrolytic graphite filter after the sample. The single crystals were mounted in closed-cycle refrigerators which allowed us to perform the measurements over a wide temperature range 10 - 300 K. The experiments for scattering in the $(0,k,l)$ zone were performed. As described in Ref. there are three different values for the lattice constant $c$. Since we will mainly show the magnetic and structural properties in the chain, $c_{\text{chain}}$ will be used to express Miller indices.

III. MAGNETIC EXCITATIONS

Figure 1b shows the temperature dependence of magnetic susceptibility parallel to the $c$ axis in single crystals of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0, 0.25,$ and $1.0$). Since the spin gap originating from the ladder has a large value of $\sim 400$ K, the susceptibility below room temperature comes predominantly from the chain. The susceptibility in Sr$_{14}$Cu$_{24}$O$_{41}$ shows a broad peak around $80$ K and the Curie-Weiss tail can be seen at low temperatures. The Curie-Weiss term is increased and the broad peak is shifted to lower temperature with yttrium substitution.

We show in Fig. 2 inelastic neutron scattering spectra at $T=10$ K observed at $(0,3,0.085)$, $(0,3,0.17)$, and $(0,3,0.25)$ in single crystals of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0$ and $0.25$). Note that the indices correspond to $(0,3,0.12)_{\text{ladder}}$, $(0,3,0.24)_{\text{ladder}}$, and $(0,3,0.36)_{\text{ladder}}$, respectively. Two sharp, intense inelastic peaks are observed in Sr$_{14}$Cu$_{24}$O$_{41}$. The peaks are the sharpest at $(0,3,0.17)$ since the resolution ellipsoid is almost parallel to the dispersion curve so that the focusing effect is expected. Thus, the intrinsic linewidth of the inelastic peaks we observed here is almost resolution-limited. Note that in the previous paper we reported the constant-$Q$ scans at $(0.3,-L)$ since the focusing condition was different for the spectrometer. The inelastic peak positions slightly change with $Q$ which follows the $\omega$-$Q$ dispersion relation as shown in the inset of Fig. 2. One puzzling feature is the presence of two excitations originating from the chain. The presence of two peaks could be due to the anisotropy in fluctuations parallel and perpendicular to the chain direction or the presence of other interactions. In Sr$_{13.75}$Y$_{0.25}$Cu$_{24}$O$_{41}$ the linewidth of the inelastic peaks becomes broader. Whereas, the peak positions are almost unchanged. These results indicate that the dimerized state becomes unstable with yttrium substitution. As described above, the broad peak in susceptibility is shifted to lower temperature in Sr$_{13.75}$Y$_{0.25}$Cu$_{24}$O$_{41}$. This is...
probably because an increase of the Curie-Weiss tail shifts the peak to lower temperature even though the intrinsic peak position is almost unchanged.

Figure 3a shows a constant-Q scan at $T=10$ K observed at $(0.3,-0.14)$ in $\text{Sr}_{13}\text{Y}_1\text{Cu}_{24}\text{O}_{41}$. The focusing effect is also expected at $(0.3,0.14)$ as at $(0.3,0.17)$. The inelastic peaks become much broader and spread over from 6 to 13 meV. This behavior is consistent with that observed in the powder sample. Since we used a single crystal, it is also possible to measure $\vec{Q}$-dependence of the magnetic excitations. We show in Fig. 3b a constant-$E$ scan at $\Delta E=8$ meV observed at $(0.3,-L)$. A broad peak can be seen around $L_{\text{chain}}=0.25$. The peak position is similar to the position at which the correlation function $S(Q)$ shows a maximum in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ suggesting that spin correlations are unchanged with yttrium substitution.

Figure 4 shows inelastic neutron scattering spectra of $\text{Sr}_{14-x}\text{Y}_x\text{Cu}_{24}\text{O}_{41}$ ($x=0$ and 0.25) at $T=10$ K observed at $(0,0,1.1)_{\text{ladder}}$. In $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ a resolution-limited sharp peak in energy was observed around 12 meV. Although the index at which the dispersion curve has a minimum corresponds to the ladder, this peak does not originate from the intra-ladder coupling since the spin gap energy is about 35 meV and the dispersion curve is expected to have minimum at $(0,0,L_{\text{ladder}})$ when $L_{\text{ladder}}=n+1/2$ or at $(H_{\text{ladder}},0,0)$ when $H_{\text{ladder}}=n+1/2$ ($n$: integer). In the previous paper, we speculated that the peak originates from the dimerized state in the ladder which is formed between the nearest-neighbor copper ions connected by the inter-ladder coupling, although the number of the dimers are considered to be small. Very recently Mikeska and Neugebauer showed that the spin gap due to the inter-ladder coupling should be much larger than 12 meV. Then the dimerization originating from the inter-ladder coupling would be realized if a local distortion occurs due to the localized holes. They also showed that the excitation at $(0,0,1)_{\text{ladder}}$ is explained with the theory of non-magnetic impurities in decoupled ladder. The sharp $(0,0,1)_{\text{ladder}}$ peak in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ becomes much broader in $\text{Sr}_{13.75}\text{Y}_{0.25}\text{Cu}_{24}\text{O}_{41}$. This strongly suggests that the excitation is closely related with the hole in the ladder, which probably couples with copper spin to form a singlet as in the chain and acts as a non-magnetic impurity.

## IV. STRUCTURAL CHANGE IN THE CHAIN

Figure 5 shows the unusual temperature dependence of the nuclear Bragg peak intensity originating from the chain for different values of $x$. For $x=0$ (Fig. 5a) the peak intensity remains constant below 30 K and then decreases with increasing temperature above 30 K. A surprising behavior was observed upon yttrium substitution. In $\text{Sr}_{13.9}\text{Y}_{0.1}\text{Cu}_{24}\text{O}_{41}$ (Fig. 5b) the intensity decreases up to 60 K and increases above 60 K with increasing temperature. In $\text{Sr}_{13.75}\text{Y}_{0.25}\text{Cu}_{24}\text{O}_{41}$ (Fig. 5c) the intensity remains constant below 30 K and then increases above 30 K. Other nuclear Bragg peaks from the chain also show fairly large temperature and yttrium concentration dependence. In $\text{Sr}_{13.9}\text{Y}_{0.1}\text{Cu}_{24}\text{O}_{41}$ for example, the intensity of the $(0,2,2)$ Bragg peak shows similar temperature dependence as that of $(0,0,2)$. The intensity at $(0,0,4)$ decreases with increasing temperature. On the other hand, the intensity at $(0,1,1)$ slightly decreases with increasing temperature.

Since ferromagnetic long-range ordering was observed in $\text{La}_6\text{Ca}_8\text{Cu}_{24}\text{O}_{41}$ which has no holes, the $(0,0,2)$ Bragg peak in $\text{Sr}_{14-x}\text{Y}_x\text{Cu}_{24}\text{O}_{41}$ might originate from a ferromagnetic ordering in the chain. The x-ray diffraction experiments were performed in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ to clarify this point. The results revealed that the $(0,0,2)$ Bragg peak intensity shows the same temperature dependence as in Fig. 5a, indicating that the Bragg intensity is nuclear in origin.

No major change of the intensity of the nuclear Bragg peak from the ladder was observed for $10<T<300$ K.

## V. DISCUSSION

We observed a drastic change of magnetic and structural properties with yttrium substitution. If the holes preferably exist on the chain, one can estimate that the number of holes $N_h$ in the chain is 60% of the copper ions in the chain in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$. Whereas, in $\text{Sr}_{13.75}\text{Y}_{0.25}\text{Cu}_{24}\text{O}_{41}$, $N_h$ in the chain is estimated to be 57.5% of the Cu ions in the chain. The change of $N_h$ is only 4.2%. It is surprising that such a small change of $N_h$ would affect the magnetic and structural properties so drastically.

Our motivation to perform the experiment using a sample with low yttrium concentrations is to clarify how the magnetic and structural properties change with the number of holes $N_h$ in the chain. Then number of the dimers which are formed between copper spins which are separated by 4 times the distance between n.n. copper ions is expected to be decreased. With further yttrium substitution, the dimers which are formed between n.n. copper spins are expected to appear. Then we expect that the
scattering around $L_{\text{chain}}=1/8$ will first disappear and then the scattering around $L_{\text{chain}}=1/2$ appear with increasing yttrium concentration.

As shown in Fig. 2, the linewidth in energy becomes broader with slight yttrium substitution. However, the integrated intensities of the inelastic peaks over energy are just slightly decreased around $L_{\text{chain}}=1/8$ and 1/4. Furthermore, we did not observe distinct inelastic peaks around $L_{\text{chain}}=1/2$ in Sr$_{13.75}$Y$_{0.25}$Cu$_{24}$O$_{41}$ or in Sr$_{13}$Y$_{1}$Cu$_{24}$O$_{41}$. Thus, the inelastic peaks become broader only in energy but not in momentum space, i.e. $S(Q)$ is unchanged. This implies that the dimerized state becomes unstable but the spin correlations are unchanged with yttrium substitution. These results are consistent with the speculation on the dimerized state in Ref. 14 that the dimerized state in the chain becomes unstable because the reduction of the holes makes the ferromagnetic nearest-neighbor interactions more dominant and the antiferromagnetic further-neighbor interaction less dominant. This is deduced, in part, from the fact that La$_6$Ca$_6$Cu$_{24}$O$_{41}$ shows a ferromagnetic long-range order in the chain. The remaining puzzle is why the scattering around $L_{\text{chain}}=1/8$ is not suppressed with increasing yttrium concentration.

We also observed that when yttrium is lightly substituted for strontium (x ≤0.25), the gap energies are almost unchanged. With further yttrium substitution (x=1.0), the excitation energy is decreased. This suggests that the exchange interaction between the spins which form the dimer is mediated by the hole at the oxygen site and that the hole probably makes the interaction longer-ranged due to the hopping mechanism. The longer-ranged exchange interaction becomes weaker when $N_h$ is reduced.

We now discuss the structural change in the chain as observed by the changes in nuclear Bragg peak intensities (Fig. 5). It is known that the lanthanide or calcium substitution for strontium affects the crystal structure. Adjacent chains are staggered in Sr$_8$Y$_2$Cu$_{25}$O$_{41}$, La$_6$Ca$_6$Cu$_{23}$O$_{41}$, and Sr$_8$Ca$_6$Cu$_{24}$O$_{41}$ (Fig. 6a-i), whereas the chains of Sr$_{14}$Cu$_{23}$O$_{41}$ slightly shift alternately along the c axis (Fig. 6a-ii). However, the structure of the ladder and strontium layers does not change with the lanthanide or calcium substitution. This suggests that the chains are almost independent of the strontium and ladder layer, which form a solid structure, and movable along the c axis relatively. In order to explain the temperature and yttrium substitution dependence of the nuclear Bragg intensity from the chain we performed a qualitative model calculation. Due to the small number of the Bragg reflections purely from the chain, it is difficult to determine the structure of the chain quantitatively. In Fig. 6a we show simple models which describe the shift of the copper ions. We also assumed that both the copper and oxygen ions shift along the c axis synchronously. The only parameter is a deviation $\delta$ along the c axis. $\delta$ equals to 0.33 at room temperature in Sr$_{14}$Cu$_{24}$O$_{41}$. Figure 6b shows the calculated intensity of (0,0,2) when $\delta$ is varied from 0 (Fig. 6a-i) to 0.5 (Fig. 6a-iii). In the calculation the contribution from the copper and oxygen in the chain was considered. The calculated intensity has maxima at $\delta=0$ and 0.5 and a minimum at $\delta=0.25$. We try to explain the experimental results based on this model. Since the scattering intensity of (0,0,2) observed in the $x=0$ sample increases with decreasing temperature, $\delta$ should become larger monotonically at lower temperature. To explain the temperature dependence of the nuclear Bragg intensity in Sr$_{13.9}$Y$_{0.1}$Cu$_{24}$O$_{41}$, $\delta$ should be slightly below 0.25 at room temperature and increase with decreasing temperature. In Sr$_{13.75}$Y$_{0.25}$Cu$_{24}$O$_{41}$ $\delta$ should be further decreased. This yttrium substitution dependence of $\delta$ is consistent with the fact that adjacent chains in Sr$_8$Y$_2$Cu$_{25}$O$_{41}$ are staggered ($\delta=0$) as described above. This simple model also explains the temperature dependence of the Bragg intensity at (0.1,0), (0.2,2), and (0.0,4). As shown in Fig. 5, the observed intensity has a finite value at the temperature where the intensity shows a minimum. The minimum value of the calculated intensity should become zero as in Fig. 6b. This is probably due to higher-order neutrons or a small distortion in the chain. We also calculated the intensity by assuming lattice distortions which could cause the spin dimerization in the chain. The intensity calculated at (0,0,2) is decreased when the lattice distortions are introduced, which is inconsistent with the experimental results in Sr$_{14}$Cu$_{24}$O$_{41}$.

In summary, we have studied magnetic and structural properties of the chains in Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ (0≤x≤1). We observed that when yttrium is substituted for strontium, strong and sharp magnetic inelastic peaks which originate from the dimerized state in the chain become broader. The peaks become broader only in energy but not in momentum space. This means that the dimerized state becomes unstable but the spin correlations are unchanged with yttrium substitution. It was also observed that nuclear Bragg peak intensities originating from the chains show strong temperature and yttrium concentration dependence. We proposed a model that the chains shift along the c axis with temperature and yttrium substitution to explain these behaviors.

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FIG. 1. (a) The chain and the ladder of copper ions in Sr$_{14}$Cu$_{24}$O$_{41}$, respectively. Filled circles represent copper atoms and open circles oxygen atoms. The dashed rectangles represent the universal unit cell in the (010) crystallographic plane. Here, $c_{\text{chain}}$ and $c_{\text{ladder}}$ represent the lattice constant $c$ for the subcells which contain the chain and the ladder, respectively.

(b) Temperature dependence of magnetic susceptibility with the external magnetic field parallel to the $c$ axis in single crystals of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0$, 0.25, and 1.0).

FIG. 2. Inelastic neutron scattering spectra of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0$ and 0.25) at $T=10$ K observed at (0,3,0.085), (0,3,0.17), and (0,3,0.25). The solid and broken lines are the results of fits to two Gaussians. The factor for the sample volume, which was estimated from phonon intensities, is corrected. The inset shows the measured dispersion (ref. 12) as a function of chain index $L_{\text{chain}}$ in Sr$_{14}$Cu$_{24}$O$_{41}$.

FIG. 3. Inelastic neutron scattering spectra of Sr$_{14}$Y$_1$Cu$_{24}$O$_{41}$ at $T=10$ K. A constant-$Q$ scan observed at (0,3,-0.14) (a) and a constant-$E$ scan at $\Delta E=8$ meV (b). The solid lines are guides to the eye.

FIG. 4. Inelastic neutron scattering spectra of Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0$ and 0.25) at $T=10$ K observed at (0,0,1.1)$_{\text{ladder}}$. The solid and broken lines are the results of fits to single Gaussian. The factor for the sample volume is corrected.

FIG. 5. Temperature dependence of the peak intensities measured at (0,0,2) in Sr$_{14-x}$Y$_x$Cu$_{24}$O$_{41}$ ($x=0$, 0.10, and 0.25).

FIG. 6. (a) A proposed model for the structural change in the chain. Open circles represent the copper ions. Note that the oxygen ions are not shown here. (b) The calculated intensity of (0,0,2) as a function of $\delta$. 
Fig. 1 Matsuda et al.
Sr$_{14-x}$ Y$_x$ Cu$_{24}$ O$_{41}$

CHAIN
(0,3,0.085)

- - $x=0$
- - $x=0.25$

Intensity (counts / 5min.)

E (meV)

(0,3,0.17)

L$_{chain}$

(0,3,0.25)

Fig. 2 Matsuda et al.
Fig. 3 Matsuda et al.
Fig. 4 Matsuda et al.
Fig. 5 Matsuda et al.
Fig. 6 Matsuda et al.