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Neutron Scattering Study in the Spin-1/2 Ladder System: $Sr_{14}Cu_{24}O_{41}$

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Inelastic neutron scattering measurements have been performed on the S=1/2 quasi-one-dimensional system $Sr_{14}Cu_{24}O_{41}$, which has both simple chains and two-leg ladders of copper ions. We have observed that both the chain and the ladder exhibit a spin gap, which originate from a dimerized state.
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1. INTRODUCTION

Recently, a spin gap has been found in the two-leg spin ladder system $SrCu_2O_3$.[1] The property of the spin gap is interesting from the view point of quantum phenomena in a low-dimensional (between 1 and 2) Heisenberg antiferromagnet. This compound have also attracted many researchers since superconductivity is expected in the carrier doped spin ladder system based on the t-J model.[2]

The structure of $Sr_{14}Cu_{24}O_{41}$[3,4] consists of two unique subcells as shown in Fig. 1. 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. We have performed inelastic neutron scattering experiments to study the dynamical magnetic properties of $Sr_{14}Cu_{24}O_{41}$.

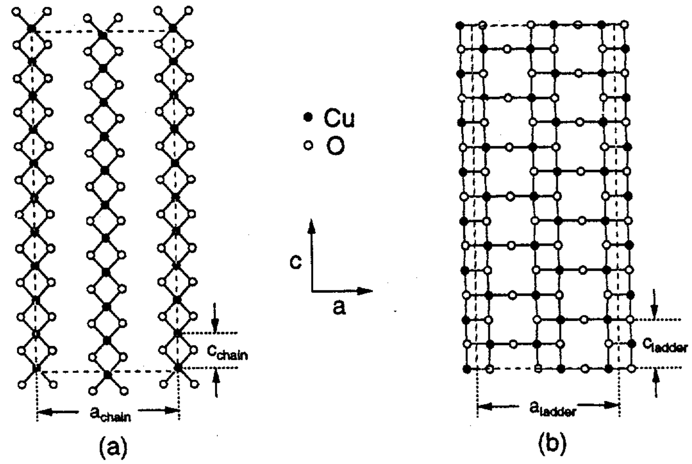

Fig. 1. The chain (a) and the ladder (b) of copper ions in $Sr_{14}Cu_{24}O_{41}$. Filled circles represent copper atoms and open circles oxygen atoms. The dashed rectangles represent the universal unit cell in the (010) crystallographic plane. Here, c_{chain} and c_{ladder} represent the lattice constant c for the subcells which contain the chain and the ladder, respectively.

2. EXPERIMENTAL METHOD

The single crystal ($\sim \phi 5$ mm$\times$30 mm) of $Sr_{14}Cu_{24}O_{41}$ were grown using a traveling solvent floating zone (TSFZ) method at 3 bar oxygen atmosphere.

The magnetic susceptibility was measured with a SQUID magnetometer (Quantum Design's MPMS2).

The inelastic neutron scattering experiments were carried out using the H7 and H8 triple-axis spectrometers at the High Flux Beam Reactor at the Brookhaven National Laboratory.

3. RESULTS

The results of magnetic susceptibility measurements on $Sr_{14}Cu_{24}O_{41}$ have shown a broad peak below 100 K.[5–8] Figure 2 shows the temperature dependence of magnetic susceptibility parallel to the c axis in a single crystal of $Sr_{14}Cu_{24}O_{41}$. The susceptibility decreases with increasing temperature and shows a minimum around 20 K and a broad peak around 80 K. The broken line in Fig. 2 shows the susceptibility after the Curie-Weiss term, which comes from paramagnetic impurities, was subtracted. The susceptibility becomes almost zero below 20 K. As is discussed by Matsuda and Katsumata,[8] the nearly 180° Cu-O bondlength parallel and perpendicular to the ladder of the two-leg ladders in $Sr_{14}Cu_{24}O_{41}$ are close to the corresponding values in $SrCu_2O_3$.[9] 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 mea-

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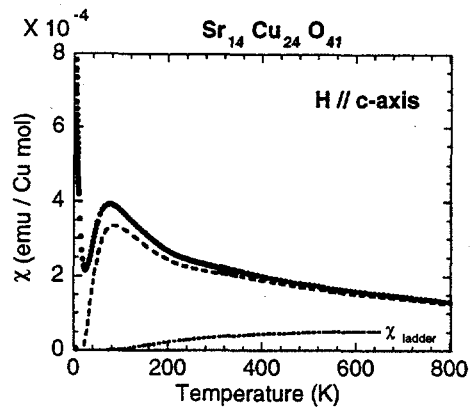

Fig. 2. Temperature dependence of magnetic susceptibility with the external magnetic field parallel to the c axis in $Sr_{14}Cu_{24}O_{41}$. The closed circles represent the observed data. The broken line represents the susceptibility after the Curie-Weiss term was subtracted. The dotted line represents the susceptibility in $SrCu_2O_3$.

surements and 680 K from NMR measurements[1]). The dotted line shows the susceptibility in $SrCu_2O_3$,[1] suggesting that the magnetic susceptibility at low temperatures comes mostly from the chain in $Sr_{14}Cu_{24}O_{41}$.

We show in Fig. 3(a) inelastic neutron scattering spectra at T=8.5 K at (0,0,1.1) and (0,3,-0.25) in $Sr_{14}Cu_{24}O_{41}$.[10] Two sharp, intense inelastic peaks are observed at (0,3,-0.25), whereas a rather weak inelastic peak is observed at (0,0,1.1).

Figure 3(b) summarizes the results of the inelastic measurements along the chain and ladder directions in $Sr_{14}Cu_{24}O_{41}$.[10] We observed a complicated ω-q dispersion relation. Here, L_{ladder} represents the Miller indices reciprocal lattice units for the ladder ($2\pi/c_{ladder}$) and L_{chain} for the chain ($2\pi/c_{chain}$). Excitations are found for L_{ladder} <0.5 and around L_{ladder}=1. We have confirmed that there is no dispersion along the a and b axes for the scattering at L_{ladder} <0.5, and no dispersion along the b axis and a weak dispersion along the a axis for the scattering around L_{ladder}=1. The positions where the dispersion curves show minima correspond to L_{chain}=1/8 and 1/4 and L_{ladder}=1. This means that the scattering below L_{ladder}=0.5 originates from the chain and that the scattering around L_{ladder}=1 from the ladder. The intensity of the constant-Q scan integrated over energy are also shown in Fig. 3(b). The intensity from the chain is about factor of 4 stronger than that from the ladder. The intensity has maximum around L_{chain}=1/4 and L_{ladder}=1.

One puzzling feature is the presence of two excitations originating from the chain as shown in the spectra of Fig. 3(a) and the dispersion curve of Fig. 3(b). The presence of two peaks could be due to the anisotropy in

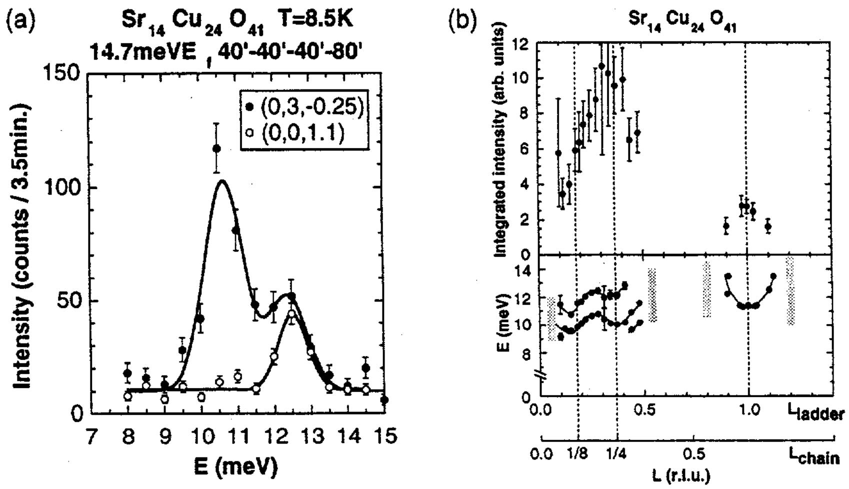

Fig. 3. (a) Inelastic neutron scattering spectra of $Sr_{14}Cu_{24}O_{41}$ at T=8.5 K observed at (0,0,1.1) and (0,3,-0.25). The solid lines are the results of fits to single Gaussian for (0,0,1.1) and two Gaussians for (0,3,-0.25). (b) The dispersion curves and the integrated intensity over energy as a function of L in $Sr_{14}Cu_{24}O_{41}$. Here, L_{ladder} represents the reciprocal lattice unit for the ladder and L_{chain} for the chain. The solid lines are guides to the eye. The shaded rectangles mean that the peak positions are difficult to be determined in these areas.

fluctuations parallel and perpendicular to the chain direction or the presence of other interactions. More experimental work is required to clarify its origin.

4. DISSCUSSION

We have observed that spin gaps exist both in the chain and the ladder. The spin gap in the chain originates from a dimerized state.[8] The spin gap in the ladder probably due to a dimerized state.[10] We now discuss the origin of the dimerized state observed in $Sr_{14}Cu_{24}O_{41}$. First, we discuss about the dimerized state in the chain. We have found from the present study that the magnetic excitations have long-range correlations; two and four times the distance between Cu^{2+} spins. If we have two neighboring Cu^{2+} spins coupled antiferromagnetically to form a singlet, we expect to see a gap at L_{chain}=1/2. However, the observed $\omega - q$ dispersion curve shows a minimum at L_{chain}=1/4 where the intensity is also a maximum. This difference suggests that there is an additional doubling along the chain direction. One possible explanation for this would be the one based on the competing nearest-neighbor and next-nearest-neighbor interactions models,[11,12] which predict incommensurate spin correlations. As mentioned above, in order to fulfill the requirement for stoichiometry, either some of the Cu ions should be trivalent or holes are created at the oxygen sites. It is believed that creating a Cu^{3+} in oxides costs larger energy than putting a hole into oxygen. A bond-valence-sums calculation has shown that Cu^{3+} ions (in other words, holes) preferably exist in the chain.[13] One can estimate that the number of holes in the chain is 60 % of the Cu ions in the chain in $Sr_{14}Cu_{24}O_{41}$. Since $Sr_{14}Cu_{24}O_{41}$ is highly insulating, the holes are considered to be localized at oxygen sites. However, hopping of these holes from one oxygen site to the other is possible. This hopping mechanism will make the exchange interactions between Cu^{2+} spins longer-ranged. It is noted that Curie-type susceptibility at low temperatures should increase considerably if the isolated Cu^{2+} ions are produced. However, the magnetic impurity density, calculated from the Curie constant assuming Cu^{2+} as impurities, is estimated to be only ~1.4 %.[8] Therefore, it is expected that most of the Cu^{2+} ions in the chain form dimers.

Another possibility to explain the magnetic excitations with longer-ranged correlations is the following. We consider the case where the holes with S=1/2 are coupled with Cu^{2+} with S=1/2 next to them and form non-magnetic state. Then the dimers can be formed between spins which are separated by 2 and 4 times the distance between the nearest-neighbor copper ions in the chain. In this case, the $\omega - q$ dispersion relation will show minima at the positions which correspond to L_{chain}=1/8 and 1/4. A puzzle remaining in the two cases mentioned above is why we do not see additional scattering from the chains at higher-order positions such as L_{chain}=3/4.

Next, we discuss the dimerized state in the ladder. As is described above, it is expected that the spin gap energy which originates from the intra-ladder coupling is above 35 meV. In this case, because of the antiferromagnetic coupling the dispersion curve is expected to have minima at $(0,0,L_{ladder})$ when $L_{ladder}=(n+1)/2$ or at $(H_{ladder},0,0)$ when $H_{ladder}=(n+1)/2$ (n: integer). The former corresponds to the dimers which are formed along the c axis and the latter along the a axis. However, we have observed minima of the dispersion at $(0,0,L_{ladder})$ where $L_{ladder}=2n+1$ (n: integer) or at $(H_{ladder},0,0)$ where $H_{ladder}=2n+1$ (n: integer). Therefore, the dimerized states, which we observed in the ladder, are formed between the nearest-neighbor copper ions which are connected by the inter-ladder coupling. Since the intensity is much weaker than that from the chain, the dimers are considered to be dilute. If we assume that there are no holes and that all the copper sites have spins, the concentration of the copper spins which contribute to the inter-ladder dimers in the ladders become ~25 % from an estimation using the integrated intensity shown in Fig. 3(b). This concentration is the upper limit. The inter-ladder dimers may be formed between spins which cannot form the intra-ladder dimer. The inter-ladder coupling constant is estimated to be antiferromagnetic and have a value of ~11 meV. This means that the ratio between the inter-ladder (~11 meV) and the intra-ladder (~100 meV) interactions is ~0.1.

In conclusion, inelastic neutron scattering measurements have been performed on the S=1/2 quasi-one-dimensional system $Sr_{14}Cu_{24}O_{41}$, which has both simple chains and two-leg ladders of copper ions. Strong magnetic inelastic peaks, which originate from the simple chains, have been observed at 9~14 meV. These excitations have been confirmed to originate from a dimerized state of the chain. The dimers are formed between spins which are separated by 2 and 4 times the distance between the nearest-neighbor copper ions in the simple chain. In addition, a weaker magnetic inelastic peak around 11 meV, which originates from a dimerized state in the ladder, has also been observed. These dimers are formed between the nearest-neighbor copper ions which are connected by the inter-ladder coupling.

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