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NQR in $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ under high pressure (1.9GPa)

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Nuclear quadrupole resonance (NQR) under steady pressure (1.9GPa) was performed on $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ where Li substitution causes metal ($x = 0$)-insulator ($x = 0.5$) transition. Two kinds of Cu-NQR signals corresponding to both states have been observed at the intermediate amount of x. Effect of the pressure was investigated for $x = 0.25$ and 0.3 samples. Possible Li-distribution pattern was proposed for the $x = 0.3$ sample.

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1. INTRODUCTION

Li substitution for Cu in LSCO system is one of the interesting problems in that the in-plane substitution causes metal-insulator transition and brings holes in CuO_2 plane like Sr substitution. Effect of the Li substitution has been mainly investigated in the end material La_2CuO_4. The main interest is whether Li-induced holes delocalize in a wide range forming the singlet with Cu ions. Antiferromagnetic long-range order (AFLRO) is lost by only 3% Li substitution, which suggests that degree of hole delocalization is high[1]. The effect on crystal structure or lattice parameters has been studied and similarities to the Sr substitution has been pointed out [2]. The susceptibility decreases with increasing Li concentration and becomes diamagnetic at 50% Li substitution[1,2]. The long-range ordering of Cu and Li cations has been observed at this concentration[3] and confinement of the singlet to CuO_4 plaquette is suggested. However, magnetic excitation gap estimated from nuclear spin-lattice relaxation is much smaller than the estimation between the singlet and the lowest-lying triplet[4].

Similar situation also holds for $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$, however, superconductivity is realized at lightly Li doped region instead of the AFLRO. One of the interesting features in this system is coexistence of two kinds of Cu-NQR signals at $x = 0.2 \sim 0.3$: One is a broad signal and the local environment is similar to that of the metallic state ($x = 0$), the other is a sharp one and is the same observed in $La_2Cu_{1-x}Li_xO_4$. Rykov *et. al.* reported that the former signal is hardly observed in $x = 0.25$ sample whereas both signals are observed in $x = 0.2$ sample[1]. Only 5% excess Li substitution induces qualitatively discontinuity in the NQR spectra. $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ is expected to locate on very critical position in the microscopic viewpoint. Thus small external action would give rise to qualitative variation in the NQR spectra. In the present work, we measured NQR spectra in $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ ($x = 0.25$ and 0.3) under steady high pressure (1.9GPa) and compared with those at 0.1GPa.

2. Experimental Equipment

Steady high pressure was retained by mounting a cryostat equipped with a piston and a cylinder on a 100t press and controlling pressure during the experiments. Powder samples plunged into liquid medium were closed by ϕ12.7 Teflon cap and Cu-Be plug. The schematic figure is shown in Fig. 1.

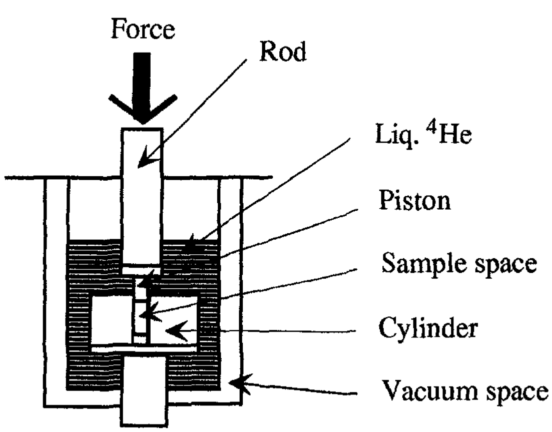

Fig. 1. Schematic figure of the cryostat mounted on a 100t press.

3. Experimental Results and Discussion

NQR spectra were measured by using a conventional spectrometer. The NQR spectra for $x = 0.25$ sample at 0.1 GPa are shown in Fig. 2(a). A broad signal around 37MHz corresponds to that of the superconductive state ($x = 0$) as is mentioned in the introduction. Although the material is an insulator as a bulk since the Li concentration is high, there still exists the environment of electric field gradient similar to the superconductive state in the microscopic viewpoint. The broad signal observed even for the $x = 0.25$ sample completely disappears for $x = 0.3$ sample as is shown in Fig. 3(a). The concentration where the broad signal disappears is quantitatively 5% larger than Rykov *et. al.*'s results [1]. In any case, the broad signal completely disappears at $x = 0.3$.

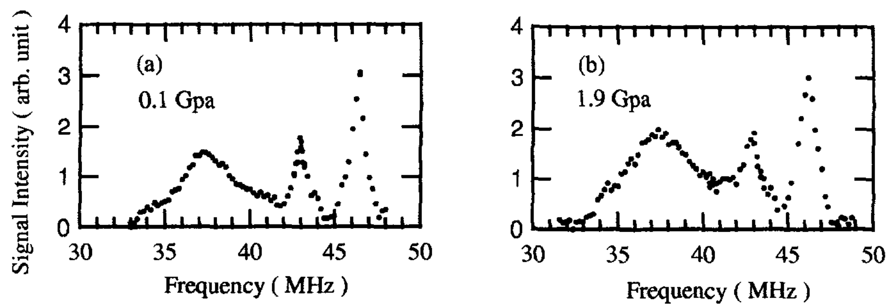

Fig. 2. NQR spectra for $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ ($x = 0.25$) at 4.2K

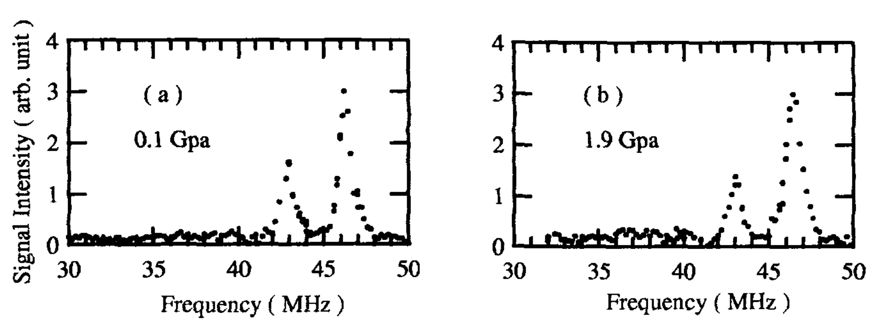

Fig. 3. NQR spectra for $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$ ($x = 0.3$) at 4.2K

NQR spectra at 1.9 GPa for the $x = 0.25$ and $x = 0.3$ samples are shown in Figs. 2(b) and 3(b), respectively. No drastic change was observed by applying pressure for the $x = 0.3$ sample except for the broadening of the linewidth. The linewidth of ^{63}Cu at 1.9GPa is 1.0MHz and is 0.2MHz broader than that at 0.1GPa. However, relative intensity of the broad signal to the sharp one changed 35% by applying pressure for the $x = 0.25$ sample.

Hole doping gives the same effect on lattice parameters irrespective of the Li substitution or the Sr substitution[2]. The lattice parameters of the CuO_2 plane shrink 4.38×10^{-3} Å for 5% hole doping, whereas the parameter for c-axis increases by the Li substitution and the rate depends on the Sr concentration. Effect of pressure on lattice is estimated from the elastic constant, $\kappa = -\frac{1}{L}(\frac{\partial L}{\partial p})$ where L is a lattice parameter. The parameters have been estimated to be 2.29×10^{-3} and -2.21×10^{-3} (GPa^{-1}) for the CuO_2 plane and c-axis, respectively[5]. Then, the lattice is expected to shrink 0.435% under the pressure of 1.9GPa for both a- and b-axes. The shrink corresponds to 5% hole doping [3]. If the shrink by the Li substitution is equivalent to that by applying pressure, the NQR spectra for the $x = 0.25$ sample at 1.9GPa should be the same with those for the $x = 0.3$ sample at 0.1GPa. However, the broad peak for the $x = 0.25$ sample does not disappear by applying pressure. Therefore, such a assumption based solely on the lattice parameters is not realized.

The present experiments suggest that the local distance of Cu-O or Li-O is rather important than the lattice parameters. Li ions occupy the Cu sites in the CuO_2 plane and break the network of Cu-O. It is known that the position of oxygen between Li and Cu ions is close to Cu ions compared with Li ions [3]. It is concluded from this fact that Li ions avoid to occupy the nearest neighbor positions each other. The fact leads to the simplest pictures of Li distribution pattern in the CuO_2 plane for $x = 1/3$ as is shown in Figs. 4(a) and (b).

Figure 4(a) represents a Li-stripe pattern where the local environment of all Cu ions is the same, namely, four nearest neighbor sites of Cu ions are occupied by two Cu ions and two Li ions. Variation of the Li-stripe pattern is also likely to realize as is shown in Fig. 4(b), however the local environment is rather close to that of Fig. 4(a) in the meaning that one of the nearest neighbor sites is Li ion. These pictures for $x = 1/3$ save to explain the drastic variation of NQR spectra at $x = 0.3$. All Cu ions possess almost the same local environment and therefore yield only one kind of NQR signals at $x = 0.3$, whereas at the concentration below $x = 0.3$ Cu ions are inevitably included whose nearest neighbor sites are occupied by four Cu ions, which gives rise to the broad signal corresponding to LSCO system.

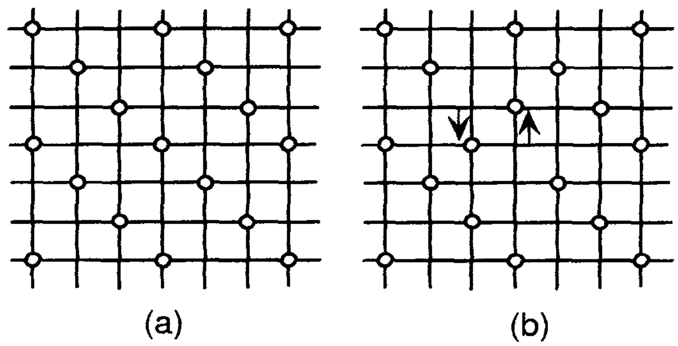

Fig. 4. Pattern of Li distribution in CuO_2 plane for 33% Li concentration. The cross represents Cu ion and open circle represents Li ion, respectively. Oxygen is not shown explicitly although it locates on the lines. (a) A Li-stripe pattern. (b) Modified pattern of figure 4(a).

4. CONCLUSION

Nuclear quadrupole resonance under steady pressure (1.9GPa) was performed on $La_{1.85}Sr_{0.15}Cu_{1-x}Li_xO_4$. Effect of the pressure was investigated for $x = 0.25$ and 0.3 samples. From the experimental results, possibility of Li stripelike pattern at $x = 0.3$ was suggested.

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