(Project use)

Experimental Report	Date of Approval 2016/8/3 Approver Ryoichi Kajimoto Date of Report 2016/8/3
Project No. 2014P0201 Title of experiment Study of structure and electronic properties of functional materials at BL01 Name of principal investigator Ryoichi Kajimoto Affiliation Japan Atomic Energy Agency	Name of instrument scientist Ryoichi Kajimoto Name of Instrument/(BL No.) 4SEASONS/BL01 Date(s) of experiment(s) 2014/5/5 11:00 – 2014/5/13 9:00 2015/3/12 11:00 – 2015/3/17 9:00

1. Outline of experimental method and results. (Experimental method and results should be reported including sample information such as composition, physical and/or chemical characteristics.)

We conducted the research subject "Study of magnetic excitations and phonons in cuprate superconductors to explore the hierarchical nature of magnetic excitations as well as the effect of spin-lattice coupling" by S2 Team in the project research group. It is well known that the hole-doped cuprate superconductors show so-called "hour-glass" magnetic excitations. The excitation appears at incommensurate positions (0.5, 0.5±δ), (0.5±δ, 0.5) in the two-dimensional reciprocal lattice at low energies. Its position gets closer to (0.5, 0.5) as energy increases, but spreads as a function of energy toward the zone boundary like an ordinary antiferromagnetic spin wave above some characteristic energy E_{cross}. Although the hour-glass magnetic excitation is regarded as one of the important ingredients for the high-T_c superconductivity, there are still several issues to be clarified. In the present study, we focused on two remaining issues for the hour-glass magnetic excitations. (1) One is to identify possible spin-charge-phonon coupled excitation. (2) The other is to characterize the true nature of the hour-glass excitations especially around E_{cross}.

(1) Study of spin-charge-phonon coupled excitation

One of the interesting features of the hour-glass excitation is that its spectral weight enhances at an energy (~15 meV) where the excitation crosses optical phonon modes. We expect it is an intrinsic phenomenon in the superconducting cuprates arising from the coupling of the magnetic excitation and the phonons. On the other hand, it is also possible that the enhancement is just a simple overlap of the two kinds of excitations. Last year (2013P0201), we have measured excitation spectra in superconducting $La_{1.925}Sr_{0.075}CuO_4$ and non-doped antiferromagnet La_2CuO_4 at 5 K over the 4-dimensional momentum-energy space by rotating the single crystals. In the superconducting sample, we have observed the enhancement in the excitation spectra at ~15 meV independent of Brillouin zones. Although it suggests that the enhancement is intrinsic to the superconducting state, to get more convincing evidence, it is important to compare the data with that at the normal state and to estimate the phonon contribution to the spectra more correctly.

1. Experimental method and results (continued)

Accordingly, this year, we performed supplementary measurements on $La_{1.925}Sr_{0.075}CuO_4$, heavily overdoped $La_{1.7}Sr_{0.3}CuO_4$, and insulating $La_{1.67}Sr_{0.33}NiO_4$ which shows stripe spin-charge ordering. All the crystals were grown by the TSFZ method. They were aligned so that the orthorhombic *a* and *c* axes are in the horizontal plane, and were rotated over 90° with 0.5° steps. Utilized incident energy was 48 meV and 55.47meV for the cuprates and nickelate, respectively.

We converted the observed excitation spectra into the dynamical spin susceptibilities $\chi''(\omega)$ for $La_{1.925}Sr_{0.075}CuO_4$ (x = 0.075) at 5 K and 200 K, and La_2CuO_4 (x = 0) at 5 K. The contribution from the simple overlap of phonon intensity in each of the spectra was removed by subtracting the excitation spectrum of $La_{0.7}Sr_{0.3}CuO_4$, because the latter compound does not show observable magnetic excitations. We obtained $\chi''(\omega)$ at several L positions to see whether it is affected by the difference in the phonon intensity as a function of L, but found that $\chi''(\omega)$ shows essentially same E (= $\hbar\omega$) dependence in all the L zones. Then, we averaged the $\chi''(\omega)$ at various L positions, which is shown in Fig. 1. For x = 0.075, thus obtained $\chi''(\omega)$ shows a clear enhancement at E ~ 15 meV in the superconducting state at 5 K, but the enhancement disappears in the normal state at 200 K. The $\chi''(\omega)$ of x = 0 does not show the enhancement even at 5 K. Furthermore, we did not observe the enhancement in $\chi''(\omega)$ of $La_{2/3}Sr_{1/3}NiO_4$ (not shown). From these observations, we conclude that the 15-meV enhancement is intrinsic to the hour-glass excitation in the superconducting state. Since the characteristic energy of the enhancement, ~15 meV, corresponds to that of several optical modes, the enhancement is possibly caused by the coupling with one of the phonon modes. One of the features in the superconducting cuprates which can connect spin and lattice degrees of freedom is that the existence of dynamical stripe-type spin-charge correlations. However, the absence of the enhancement in the excitation spectrum in stripe-ordered $La_{2/3}Sr_{1/3}NiO_4$ suggests that the static stripe correlations cannot be the origin of the enhancement. Then, we expect that the enhancement is a result of spin-charge-lattice (phonon) coupling through the *dynamical* stripe correlations. We are now performing phonon calculation to find any optical phonon mode that can couple the dynamical stripes and can be the origin of the 15-meV enhancement.

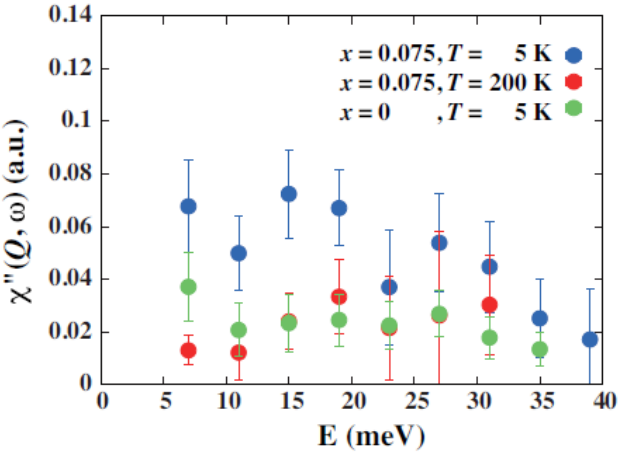

Fig. 1. Dynamical spin susceptibility $\chi''(\omega)$ for $La_{1.925}Sr_{0.075}CuO_4$ at 5 K and 200 K, and La_2CuO_4 at 5 K.

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1. Experimental method and results (continued)

(2) Study of hierarchical structures in the hour-glass magnetic excitation

Recent experimental studies suggested that the hour-glass magnetic excitation consists of two components with different origins, whose ratio varies as a function of energy. To confirm the existence of the two components, we performed inelastic neutron scattering experiments on superconducting hole-doped copper oxides $La_{2-x}Sr_xCuO_4$. In previous inelastic neutron scattering studies, it was difficult to prove the existence of the two components because of poor statistics of data. In the present study, to investigate detailed structures in the excitation spectra, inelastic scattering intensity was collected with much higher statistics than that available before, thanks to the high performance of 4SEASONS.

Measured samples are single crystals of x = 0.10 and 0.16, which were grown by the TSFZ method. The crystals were aligned so that the c axis is parallel to the incident neutron beam. The excitation spectra as functions of the two-dimensional momentum transfers and energy transfer were obtained neglecting the c^* dependence of the spectra.

Figures 2(a)-2(c) show constant-E slices of the magnetic excitation spectrum in x = 0.16, while Fig. 2(g) shows the h-E slice of the spectrum. Our high-statistics data clearly shows that the incommensurate structure in the low-energy part of the hour-glass excitation does not considerably change as a function of energy [Fig. 2(g)]. That is, the incommensurability δ of the magnetic excitation does not show energy dependence. Based on this experimental result, we modeled the excitation spectrum as a combination of two components: one is incommensurate with a fixed δ and the other is commensurate centered at (0.5, 0.5). By using this model, we successfully reproduced the observed spectrum below 60 meV as shown in Figs. 2(d)-2(f). The result of the present study provides a strong evidence of the two components in magnetic excitations of superconducting cuprates.

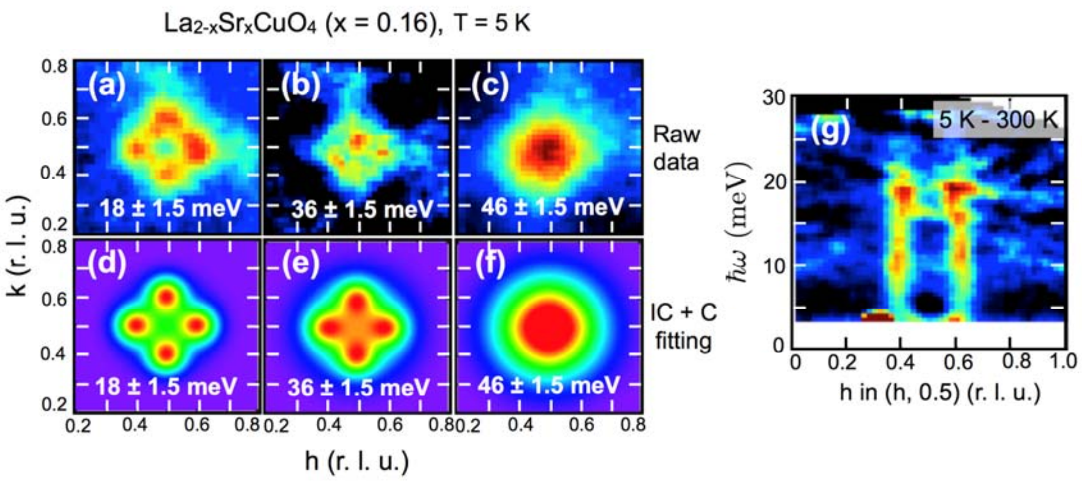

Fig. 2. (a)-(c) Constant-E slices of the magnetic excitation spectrum in $La_{2-x}Sr_xCuO_4$ (x = 0.16) at 18, 36, and 46 meV. (d)-(f) Calculated spectra assuming the combination of incommensurate and commensurate components. (g) Slice of the magnetic excitation spectrum on the (h,0.5)-E plane. The intensity at 300 K is subtracted.

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