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Measurements of two-dimensional antiferromagnetic spin wave by using chopper spectrometer installed in pulsed neutron source

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ABSTRACT

Neutron inelastic scattering measurements on the chopper spectrometer MARI installed in the pulsed neutron source ISIS observed a well defined two-dimensional antiferromagnetic spin wave up to the zone boundary of La_2NiO_4 at 124±3 meV.

I.Introduction

Low dimensional antiferromagnetism provides a wide variety of challenging issues such as Haldane's conjecture[1)], the resonating-valence-bonding state[2)] and so on, relating with quantum effects on the spin fluctuations which are enhanced due to the low dimensionality. Two-dimensional antiferromagnetism has been especially put high light after the discovery of the superconductivity in 2D copper oxides because the 2D magnetic interactions between Cu spins have been regarded as one of the candidates for the microscopic interactions to create the Cooper pairs.

Triple-axis neutron scattering measurements first found out unique 2D spin fluctuations in La_2CuO_4 [3)], a mother compound providing a framework for Tc=40 K class oxide superconductor. These techniques also revealed characteristic change of the fluctuations by carrier doping.[4)] In order to prove the correlation of the magnetism with the superconductivity, more quantitative study in a wider q-ω space is necessary because a shift of the energy specrtum in $\chi(q,\omega)$ to higher energies is expected when the superconducting state appears. However, an extention of the energy region by using triple-axis technique is very difficult.

Instead chopper spectrometer coupled with pulsed neutron spallation source has been regarded as one of the better spectrometers in high energy region compared with triple-axis one especially for low dimensional magnetic systems because of the larger detector area, the better energy-resolution and the higher signal-to-noise ratio in high energy region. Therefore pulsed neutron scattering measurements on La_2CuO_4 and the related compound are of great interest not only to study the quantum effects on the 2D spin fluctuations but also to test the capability of the chopper spectrometer. Systematic studies on 2D antiferromagnetic systems with different spin values are also important to clarify the quantum nature of spin fluctuations in 2D antiferromagnets. We believe a series of 2-1-4 compound, La_2CuO_4 with S=1/2, La_2NiO_4 with S=1 and La_2CoO_4 with S=3/2 is the most ideal system to study the spin value dependence of the magnetism.

In the present paper results of pulsed neutron inelastic scattering measurements on single crystal of 2D antiferromagnet La_2NiO_4 are reported. By using chopper spectrometer we first observed a well defined spin-wave up to the zone-boundary and determined the in-plane exchange interaction J.

II.Experimental details

Single crystals of $La_2NiO_{4+\delta}$ with average dimensions of 8 mm$^{\phi}$x 35 mm were grown by a floating zone method in air. As-grown crystals are reduced in a CO_2 mixed with 0.1% CO gas flow at 1100 C for 30 h. This heat treatment reduced the excess oxygens in the as-grown crystals probably to δ~0.02 because the 3D Neel temperature changed from 60 K to 210 K.[5] At room temperature the crystal structure is of two phases, orthorhombic and probably tetragonal phase due to the small amount of excess oxygen. Below around 150 K, the crystal trnsforms into single tetragonal phase with $P_{42/ncm}$ symmetry.

Neutron inelastic scattering measurements were carried out on the chopper spectrometers INC and MARI installed in the spallation neutron sources in the National Laboratory for High Energy Physics and in Rutherford Appleton National Laboratory, respectively. The nominal energy-resolution is about 2.5% for INC and 1% for MARI. In chopper spectrometer coupled with pulsed neutron source, the monochromated incident pulsed beam is provided by a mechanical chopper synchronized with the repetition of the neutron pulse. The energy of incident neutron beam is tunable by changing the phase and/or the frequency of the rotation of the chopper. The scattered neutrons are detected by a large number of ^{3}He detectors aligned in a wide range of scattering angles in both horizontal and vertical directions. In this report we present the data obtained by the low angle-horizontal detector bank in MARI with the scattering angles from 3° to 9°. Other data taken by INC and by vertical detectors which can be used as complementary data will be presented in a separated paper.

For single crystal measurements, crystal orientation is important to extract useful and simple information. In the present study the data were taken by two types of crystal orientations, which are described in the following.

II-1 Orientation with magnetic Bragg lines perpendicular to k_i

In this orientation, hereafter we call type-A configuration, single crystals of La_2NiO_4 were aligned in a direction with the 2D magnetic Bragg lines perpendicular and with the a-axis parallel to the incident neutron momentum k_i as shown in Fig. 1. In time-of-flight (TOF)-scans, which correspond to k_f-scans, detectors pick up magnetic intensities when the scan trajectories cross the 2D spin wave dispersion surfaces. The energy difference in the TOF-scan trajectories of different detectors at the same 2D momentum transfer q_{2D} is less than 2 % for the transferred energies larger than 50% of the incident ones. Since a matching of energy resolution is achieved in this measurement, the intensities picked up by different detectors between at 3° and 9° can be binned up without degradation of the energy resolution. Several incident nuetron energies from 70 meV to 400 meV are carefully

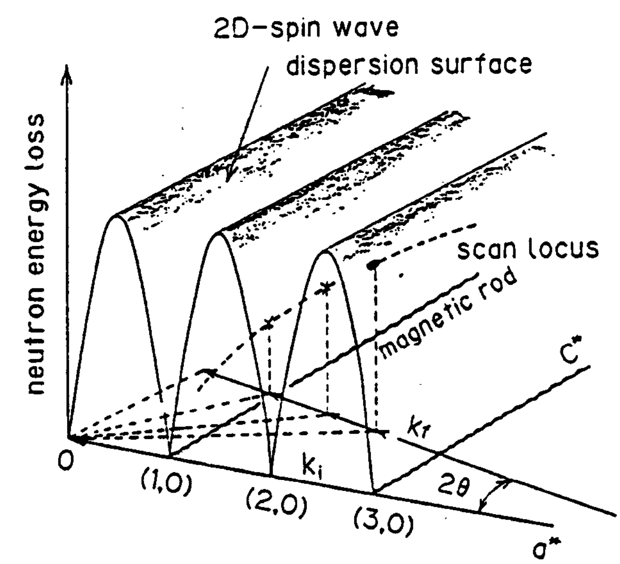

Fig. 1 Type-A orientation

selected to perform proper scans in a wide reciprocal space.

II-2 Orientation with magnetic Bragg lines parallel to k_f

In another orientation, type-B orientation, the crystals are aligned with the 2D magnetic lines parallel and with the a-axis perpendicular to k_f. In this orientation the TOF-scan corresponds approximately to constant q_{2D} scan as shown in Fig. 2. In the figure thick and fine lines denote dispersion curves and scan trajectories, respectively. With this orientation however, neutron counts of a small number of detector can be binned because the scan trajectories are extremely different for different scattering angles compared with type-A orientation.

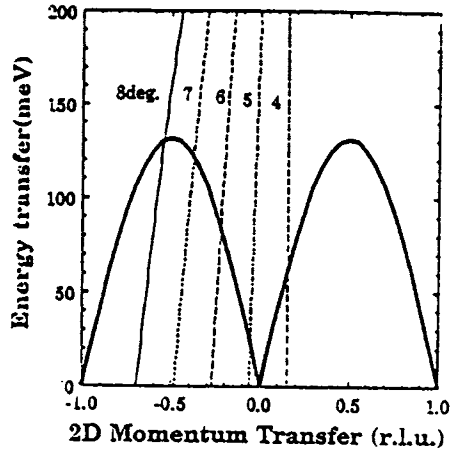

Fig. 2 Type-B orientation

III. Results and Discussion

In Fig. 3 we show a typical spectrum taken by the type-A orientation with an incident energy of 165 meV. The inserted figure shows the expanded one around 100meV. The neutron counts of the 28 horizontal detectors at both left and right hand side are binned in the figure. If observed peaks are originated by spin-wave excitations, they should be mapped on dispersion curves as well. Both TOF trajectories of different incident energies and antiferromagnetic spin-wave dispersion curves are drawn in Fig. 4. Closed circles or ellipsoids on the dispersion curves therefore, represent peak positions of spin-wave. Circles which are not mapped on dispersion curves are considered to be originated by phonon excitations. By changing the incident neutron energies we traced the spin-wave dispersion curves up to the zone-boundary at 124±3 meV. A full width at half maximum of the spin-wave peak at around 117 meV in Fig.3 is about 2.5 % of the transferred energy which is close to the energy-resolution. Due to the high energy-resolution of MARI even the low energy spin-wave peak around 40 meV exhibits a double-peak shape.

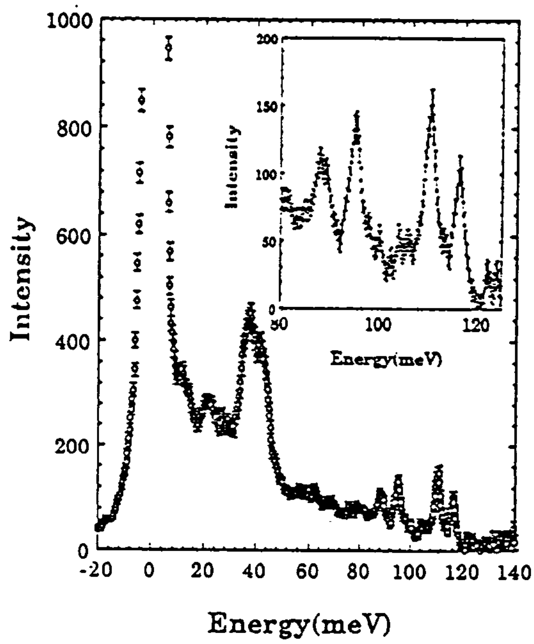

Fig. 3 A typical spectrum taken by type-A orientation

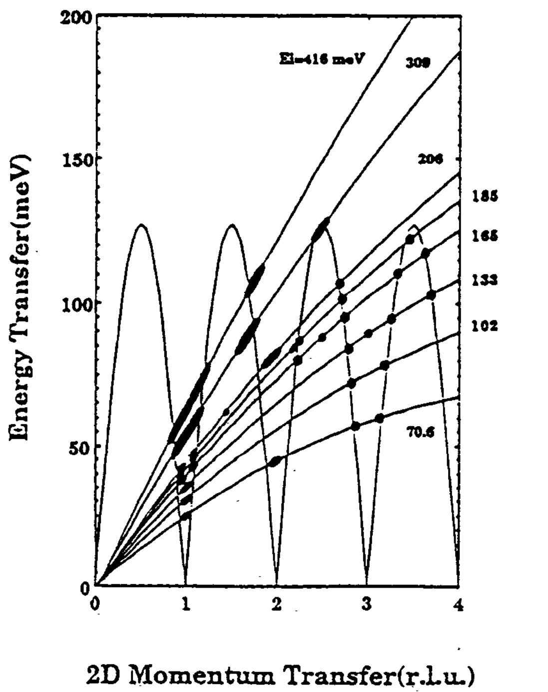

Fig. 4 Dispersion relation and scan trajectories for type-A orientation

A spectrum obtained by the type-B orientation is shown in Fig. 5 where the neutron events detected at the scattering angles between 7° and 8° are binned. Around 125 meV there is a remarkable change in the neutron intensites. As is expected from Fig. 3 and Fig. 4, this energy may correspond to the energy around the zone-boundary. However quality of the data is much worse than that of the type-A orientation due to the smaller number of detectors available in the type-B.

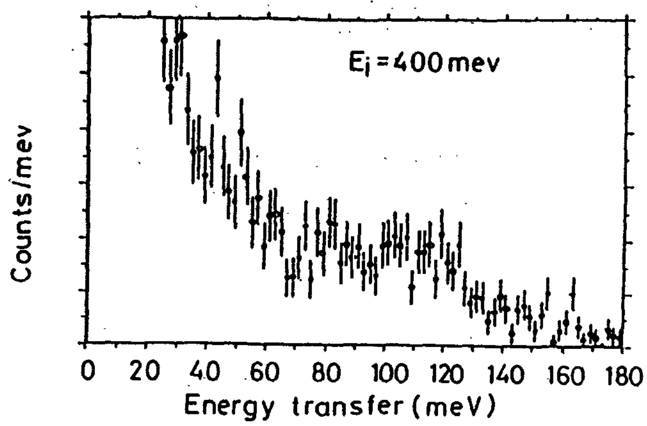

Fig. 5 A typical spectrum taken by type-B orientation

The peak positions of spin-wave observed by the type-A orientation almost perfectly sit on a simple dispersion relation for 2D square-lattice antiferromagnet,

$$\hbar\omega=4SJ\sin(\pi q_{2D}(\mathrm{r.l.u.}))$$

with an in-plane exchange constant J between Ni^{2+} spins of 31± 1 meV. This value is consistent to that of 30 meV obtained from the two-magnon peak observed by Raman measurement [6] where the peak position is predicted to correspond to 6.7J by a spin-wave theory taking into magno-magnon interactions. Therefore within the preliminary data analysis, the observed spin-wave dispersion relation in La_2NiO_4 can be described by a simple spin-wave theory.

Analyses taking into the experimental resolution function will provide more detailed imformations such as the line-widths and the intensities in the magnetic excitations. Although the obtained J value is smaller by factor about three than that is expected in La_2CuO_4, we believe the same technique which is performed here is also well applicable in La_2CuO_4.

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