（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。）

<table>
<tr><td>TOKAI CROSS Experimental Report J-PARC</td><td>提出日 Date of Report
8 November, 2012</td></tr>
<tr><td>課題番号 Project No.
2012A0144
実験課題名 Title of experiment
Molecular spin polaron with colossal magnetic moment in lightly impurity doped LaCoO3 systems
実験責任者名 Name of principal investigator
Keisuke Tomiyasu
所属 Affiliation
Tohoku University</td><td>装置責任者 Name of responsible person
Ryoichi Kajimoto
装置名 Name of Instrument/（BL No.）
4SEASONS (BL-01)
実施日時 Date and time of Experiment
21 May to 25 May and 28 October to 31 October (totally 7 days)</td></tr>
</table>

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Perovskite $LaCo_{0.99}Ni_{0.01}O_3$, $LaCo_{0.95}Te_{0.05}O_3$, and $LaCo_{0.80}Te_{0.20}O_3$

Powder form

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

I. Introduction

The perovskite-type cobalt oxide $LaCoO_3$ (Co^{3+}: d^6) is a nonmagnetic insulator with low-spin state (spin S = 0) below about 100 K. Interestingly, in the low-spin state phase in $La_{0.998}Sr_{0.002}CoO_3$ with very lightly doped holes, superparamagnetism with a colossal magnetic moment over 10 μ_B/hole is observed in magnetization measurements [Yamaguchi *et al.*, PRB (1996)]. Recently, as its origin, powder inelastic neutron scattering suggested that six Co^{3+} ions surrounding Co^{4+} (hole) change from low-spin to magnetic intermediate-spin state (S = 1), probably caused by ferromagnetic double-exchange mechanism between Co^{3+} and Co^{4+} (molecular spin polaron) [A. Podlesnyak *et al.*, PRL (2008)]. This spin polaron is interpreted as a precursor of carriers, which generate giant magnetic resistance, anomalous Hall effect, and thermopower in a semiconductor region and a ferromagnetic metal region with higher hole concentration.

Inspired by the hole-doping experiments, we have tackled the generation of intriguing phenomena with novel mechanisms utilizing the spin-state responses. Here, we measured the Ni-doped and Te-doped samples. We previously confirmed the following characters in our measurements of macroscopic properties

2. 実験方法及び結果（つづき） Experimental method and results (continued)

and synchrotron x-ray fluorescence; the Ni doping corresponds to orbital doping and induces sensitive magnetic responses, whereas the Te doping corresponds to electron doping and hardly affects both magnetism and electric conductivity.

II. Experimental method

Standard inelastic neutron scattering experiments for the powder specimens were performed using the CCR.

III. Results

Figure 1 shows the measured inelastic neutron scattering data obtained at the present lowest temperature 7 K. The left panel shows the data of Ni-doped system. The non-dispersive spin excitation mode is observed around momentum around 1.2 meV, which is very analogue with that observed in the aforementioned Sr-doped system. Therefore, the emergence of similar molecular spin state polarons is strongly suggested in the Ni-doped system as well. Unfortunately, however, we could not further explore this signal because of the insufficient detector coverage and energy matching. Thus, we proposed the continuous experiments to AMATERAS and will perform the further investigation in the 2012B period.

The middle and right panels show the data of $LaCo_{0.95}Te_{0.05}O_3$ and $LaCo_{0.80}Te_{0.20}O_3$. The flat modes are observed around 22 meV, of which the intensity increases with increasing the Te concentration. In analogy with magnetic excitations in CoO and La_2CoO_4 that have high-spin Co^{2+} with spin-orbit coupling and unquenched orbital angular momentum, these flat modes are most probably identified to be the internal first spin-orbit excitation level of Co^{2+}. Thus, our data strongly suggest that the doped electrons generate the well-defined Co^{2+} ions, not an intermediate valence between divalence and trivalence. Now based on the information, we are discussing the origin of insensitive magnetism and electronic conductivity.

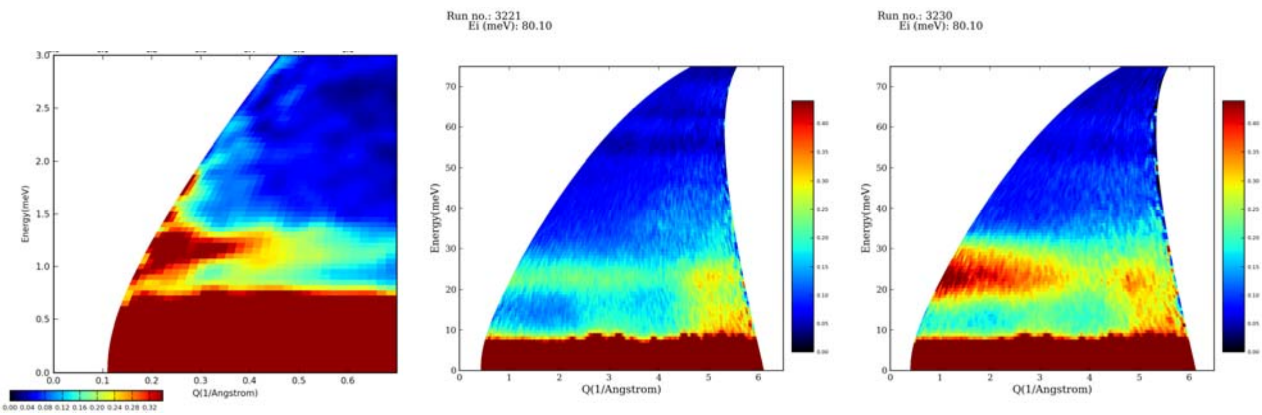

Figure 1. Measured inelastic neutron scattering data obtained at 7 K. The left, middle, and right panels show the scattering intensity distribution in momentum (*Q*) and energy (*E*) space measured on $LaCo_{0.99}Ni_{0.01}O_3$, $LaCo_{0.95}Te_{0.05}O_3$, and $LaCo_{0.80}Te_{0.20}O_3$, respectively.