


(※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。)

 MLF Experimental Report	提出日 Date of Report 2015 2/18
課題番号 Project No. 2014B0235 実験課題名 Title of experiment Determination of the crystal and magnetic structures of $S = 3/2$ classical triangular antiferromagnet β -LiCoPO ₄ 実験責任者名 Name of principal investigator Hiroyuki Yoshida 所属 Affiliation Hokkaido University	装置責任者 Name of responsible person 神山崇 装置名 Name of Instrument/(BL No.) Super High Resolution Powder Diffractometer (BL08) 実施日 Date of Experiment 16-19 Dec 2014

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.
<p>β type Lithium Cobalt phosphate (β-LiCoPO₄)</p>

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.
<p>Our study focuses on the magnetic properties of β-LiCoPO₄ which could be an ideal candidate for the $S = 3/2$ classical triangular antiferromagnet. We have already revealed that the compound shows successive magnetic transitions at $T_{N1} = 17$ K and $T_{N2} = 11$ K by means of the magnetic susceptibility and heat capacity measurements.</p> <p>We carried out neutron powder diffraction experiments on β-LiCoPO₄ at BL08 SuperHRPD. The measurements were conducted under various temperatures, $T = 5, 7, 9, 10, 11, 13, 15, 16, 17, 18, 20$ K to investigate crystal and magnetic structures of the compound below T_{N1} and T_{N2}.</p> <p>First, we successfully refined the detailed crystal structure of the material at 20 K (in the paramagnetic phase) by the Rietveld analysis using Z-rietveld program, since the Li position of the compound haven't been completely determined by XRD experiments. The structural parameters of β-LiCoPO₄ is summarized in Table 1 and the structure is illustrated in Fig. 1. We also investigated the crystal structure below magnetic transition temperatures and found that there is no structural phase transition below 20 K.</p>

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

Next, we investigated the magnetic structure of the compound. We successfully observed the magnetic Bragg reflections below T_{N1} as shown in Fig. 2. The magnetic propagation vector k_m of intermediate phase ($T_{N2} < T < T_{N1}$) is $k_m = (q, 1, 0)$ which q value changes with decreasing temperature (ex. $q = 0.28$ at $T = 13$ K). This results show that an incommensurate magnetic state is realized in the intermediate phase as illustrated in Fig. 3(a). Interestingly, we observed that a commensurate magnetic structure with $k_m = (0, 1, 0)$ is realized below the phase transition at T_{N2} as shown Fig. 3(b). In addition, we found that the two phases coexists below the lower transition temperature T_{N2} , suggesting the first-order phase transition occurs at the temperature which is consistent with the results of our previous heat capacity measurements. As we already mentioned above, there is no structural phase transition down to lowest temperature, this first-order phase transition is originated from magnetic properties of the compound.

These results obtained by the neutron powder diffraction measurements at BL08 J-PARC are quite rare and interesting. The origin of these behavior is still unclear, however we expect that these magnetic properties are attributed to the magnon Wigner crystallization mechanism proposed by some theories. To clarify the phenomena observed in our experiments, we need further analyses of the data based on theoretical suggestions.

Table 1 Structural parameter of β -LiCoPO₄

	Site	x	y	z	Occupation	U (Å ²)
Li	4c	0	0.6717	0.25	1	0.007
Co	4a	0	0	0	1	0.002
P	4c	0	0.35177	0.25	1	0.001
O1	8f	0	0.24614	0.04638	1	0.003
O2	8g	0.22827	0.46722	0.25	1	0.003

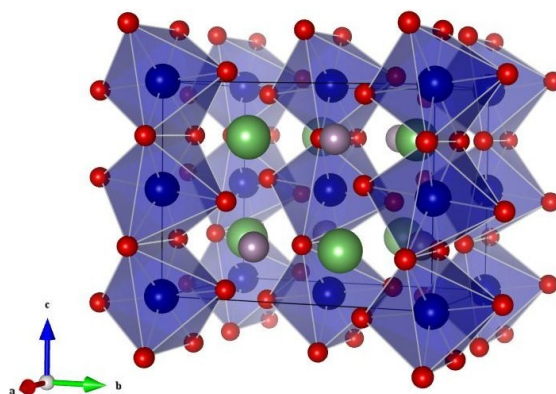


Fig. 1 Crystal structure of β -LiCoPO₄

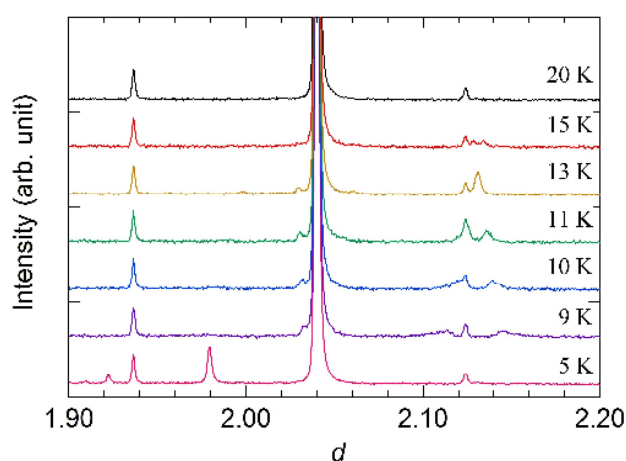


Fig. 2 Neutron diffraction pattern of β -LiCoPO₄ under various temperatures.

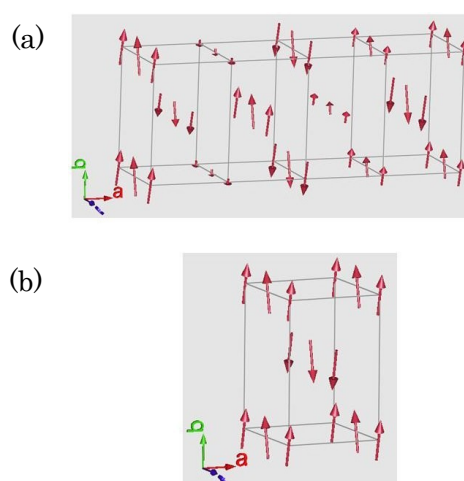


Fig. 3 Expected magnetic structure of (a) β -LiCoPO₄ at 13 K (intermediate phase) and (b) 5 K (low temperature phase).