

Guochu Deng<sup>1</sup>, Yiming Cao<sup>2</sup>, Wei Ren<sup>2</sup>, Shixun Cao<sup>2</sup>, Andrew Studer<sup>1</sup>, Nicolas Gauthier<sup>3</sup>, Michel Kenzelmann<sup>3</sup>, Gene Davison<sup>1</sup>, Kirrily Rule<sup>1</sup>, Jason Gardner<sup>4</sup>, Paolo Imperia<sup>1</sup>, Clemens Ulrich<sup>5</sup>, and Garry McIntyre<sup>1</sup>

<sup>1</sup>*ACNS, Australian Nuclear Science and Technology Organisation, Australia*

<sup>2</sup>*Department of Physics, Shanghai University, China (P.R.C)*

<sup>3</sup>*Laboratory for Scientific Developments & Novel Materials, Paul Scherrer Institut, Switzerland*

<sup>4</sup>*Neutron Group, National Synchrotron Radiation Research Center, Taiwan*

<sup>5</sup>*School of Physics, University of New South Wales, Australia*

$\text{Co}_4\text{Nb}_2\text{O}_9$  was recently reported to have large magnetoelectric coupling effect under a certain magnetic field.<sup>[1,2]</sup> This compound has a crystal structure (space group  $P-3c1$ ) derived from corundum structure and undergoes antiferromagnetic phase transition around 27K. It was previously believed that the magnetic moments of  $\text{Co}^{2+}$  order into a collinear antiferromagnetic structure in which magnetic moments are parallel to the  $c$  axis and form ferromagnetic chains with antiparallel inter-chain coupling.<sup>[3]</sup> However, the recent study has shown that this magnetic structure model is incorrect.<sup>[4]</sup> In this study, we found that the  $\text{Co}^{2+}$  magnetic moments on both Co1 and Co2 sites align in the  $ab$  plane with a non-collinear configuration. Using inelastic neutron scattering, we measured the spin wave excitation from its magnetic phase along  $(h00)$  and  $(00l)$ . A spin dynamic model proposed in this study is able to explain the observed spin dynamical behavior quite well. The nearest and next nearest neighbor interactions (NN and NNN) along the  $c$  axis are ferromagnetic. The interaction on the zig-zag ring of Co1 perpendicular to the  $c$  axis is highly frustrated while that of the zig-zag ring of Co2 is antiferromagnetic. The single ion anisotropy and Dzyaloshinskii-Moriya (DM) interaction contribute to the spin dynamics of  $\text{Co}_4\text{Nb}_2\text{O}_9$  as well. The simulated spin wave excitation by using SpinW<sup>[5]</sup> matches the experimental data very well. The DM interaction, which is most probably due to the triangle Co2-O-Co2 bond, was found to be the origin of the magnetoelectric coupling in this compound.

[1] Yiming Cao, Yali Yang, Maolin Xiang, Zhenjie Feng, Baojuan Kang, JincangZhang, Wei Ren, Shixun Cao, *J Cryst. Growth* **420**,90-93(2015)

[2] Y. Fang, et al., *Scientific Reports* **4**, 3860 (2014)

[3] E.F. Bertaut, et al., *J Phys. and Chem.of Solids* **21**, 234-251 (1961)

[4] N.D. Khanh, et al. *Phys. Rev. B* **93**,075117 (2016)

[5] S. Toth, et al. *J.Phys.: Condens. Matter* **27**, 166002 (2015)