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ACCEPTED MANUSCRIPT

Theory of magnetoelectric response in $Co_4Nb_2O_9$

Yuki Yanagi^{a,*}, Satoru Hayami^b, Hiroaki Kusunose^a

^aDepartment of Physics, Meiji University, Kawasaki 214-8571, Japan ^bDepartment of Physics, Hokkaido University, Sapporo 060-0810, Japan

Abstract

The mechanism of the observed novel magnetoelectric effects in $\rm Co_4 Nb_2 O_9$ is addressed by calculating the magnetoelectric tensor of the realistic effective model derived from the first principles calculation. The obtained magneticfield angle dependence of the electric polarization qualitatively reproduces the experimental result. It is shown that the orbital contribution dominates the magnetoelectric response and the angle dependence can be understood by the first order perturbation with respect to the spin-orbit coupling.

Keywords: Magnetoelectric effect, Co₄Nb₂O₉

1. Introduction

Strongly correlated electron systems show many intriguing physical phenomena due to the interplay between spin, charge and orbital degrees of freedom. Among them, magnetoelectric (ME) effects, i.e. magnetization induced by the external electric field or electric polarization by the magnetic field, have attracted considerable interest in many years [1–18]. The linear ME effects are characterized by the magnetoelectric tensor $\alpha_{rr'}$ (r, r' = x, y, z) which can have nonzero values with the lack of both time reversal and inversion symmetries.

 Cr_2O_3 is a prototypical example of ME materials, which shows ME effects in the antiferromagnetic (AFM) phase [19, 20]. The crystal structure of Cr_2O_3 is the corundum structure, where Cr-ions form the honeycomb lattice in the *ab* plane and the inversion center is not located on a Cr-site. Then, the collinear AFM order breaks both the time-reversal and inversion symmetries, resulting in the emergence of linear ME effects. The magnetic point group of the AFM state is $\bar{3}'m'$, where the AFM moment is directed along the *c*-axis and the ME tensor has a diagonal form. Another example of ME materials, honeycomb antiferromagnet $Co_4Nb_2O_9$, also shows ME effects below the Néel temperature as in the case of Cr_2O_3 [21], where the lattice structure is shown in Fig. 1 (a) [22, 23]. Recently, magnetic properties and ME response of $Co_4Nb_2O_9$ have been reinvestigated by several authors [24–29]. According to the neutron

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^{*}Corresponding author Email address: y_yanagi@meiji.ac.jp (Yuki Yanagi)

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