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Synthesis and physical properties of spin-1 honeycomb lattice $\text{Pb}_6\text{Ni}_9(\text{TeO}_6)_5$

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Abstract

We report the magnetic and dielectric properties of the spin-1 honeycomb lattice compound $\text{Pb}_6\text{Ni}_9(\text{TeO}_6)_5$. It crystallizes in hexagonal structure with a non-centrosymmetric space group $P6_322$. DC magnetic susceptibility, AC magnetic susceptibility, and heat capacity studies reveal a magnetic long-range-order at $T_N \simeq 24$ K with a weak ferromagnetic component. The ground state is identified to be a canted antiferromagnetic type which is driven by the Dzyaloshinsky-Moriya interaction. A change in slope in the dielectric permittivity and a sudden drop in the loss tangent at T_N implies the coupling of electric and magnetic order parameters.

Keywords: Oxide Materials, Heat capacity, Dielectric response, Magnetic measurements

1. Introduction

The study of quantum phase transition in low-dimensional spin systems has gained wide attention from both experimental and theoretical research point of views.[1] For conventional one-dimensional (1D) and two-dimensional (2D) systems, the ground state properties are well established by several numerical calculations and subsequent experimental realizations. For instance, magnetic long-range-order (LRO) is forbidden in 1D and 2D spin-1/2 Heisenberg spin systems down to zero temperature. However, 2D XY systems don't completely preclude LRO but it undergoes a Kosterlitz-Thouless transition: a topological phase transition caused by the unbinding of vortex-antivortex pairs at a finite temperature.[2, 3] Investigating such novel ground state properties are important in order to unravel the mechanism behind these phases.

Honeycomb lattices in 2D are considered to be XY model systems. In real materials, in-plane anisotropy, inter-layer coupling, competing interactions, orbital degree of freedom etc always lead to a deviation from the purely XY behaviour and

modify the ground state significantly. In certain cases, the electron or hole doping also significantly influence the ground state. Some of the interesting ground states in honeycomb lattices arising due to such effects are spin-orbital liquid in $\text{Ba}_3\text{CuSb}_2\text{O}_9$,[4, 5] exotic disorder state in $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$,[6] spin-glass behaviour in $\text{Li}_4\text{MgReO}_6$,[7] singlet state in $\text{Na}_3\text{Cu}_2\text{SbO}_6$,[8, 9] superconductivity in doped $(\text{Zr,Hf})\text{NCl}$,[10, 11] etc.

Herein, we report synthesis, magnetic, and dielectric properties of $\text{Pb}_6\text{Ni}_9(\text{TeO}_6)_5$. It crystallizes in a hexagonal structure with space group $P6_322$ (No. 182) and lattice constants $a = b = 10.257(1)$ Å, and $c = 13.554(5)$ Å.[12] There exist two inequivalent Pb sites, two inequivalent Te sites, four inequivalent Ni sites, and five inequivalent O sites in the crystal structure. NiO_6 octahedra in which Ni^{2+} exists in spin-1 are edge shared forming 2D honeycomb layers in the ab -plane, as shown in Fig. 1. The distance between Ni^{2+} ions in each hexagonal unit is 2.913 Å which facilitates uniform antiferromagnetic (AFM) nearest-neighbour (NN) interaction. Two TeO_6 units are located symmetrically above and below the center of each hexagon. Each TeO_6 unit connects all the six Ni^{2+} ions within one hexagon providing the next-nearest-neighbour (NNN) interaction path between Ni^{2+} ions. For clarity, we have

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