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## ACCEPTED MANUSCRIPT

# The regulation of magnetoelectric coupling and magnetic exchange bias effects in La doped Bi<sub>5</sub>Ti<sub>3</sub>FeO<sub>15</sub> multiferroic films

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#### Abstract

La doped Bi<sub>5</sub>Ti<sub>3</sub>FeO<sub>15</sub> (BTFO) multiferroic films with various doping concentration were synthesized by chemical solution deposition method. The structure, ferroelectric, magnetic properties, and magnetoelectric coupling effect were systemically investigated. Room temperature magnetoelectric effect was obtained. And the magnetoelectric coupling and magnetic exchange bias effects are regulated by the doping concentration of La. Such regulation originates from the evolution of magnetic structure derived from doping rare earth La. The coexistence of ferromagnetic (FM) and antiferromagnetic (AFM) orders can be controlled by the doping concentration of La. Thus, the superexchange interaction between FM and AFM orders was enhanced strongly, which contributes to the strong magnetoelectric coupling effect. This work provides an avenue on regulating the magnetic structure and magnetoelectric coupling effects.

Keywords: Magnetic materials; Dielectrics; Ferroelectrics; Magnetoelectric coupling; Exchange

bias effect

#### Introduction

Magnetoelectric effect has been intensively explored due to its fascinating physics phenomenon and potential application in low power information storage, energy harvest and smart sensor areas.<sup>1-3</sup> As single phase materials with four-layer perovskite structure, Bi<sub>5</sub>Ti<sub>3</sub>FeO<sub>15</sub> (BTFO) films have been received much attention due to their excellent multiferroic properties.<sup>4,5</sup> Some symbolic results have been achieved for BTFO multiferroic materials, such as the ferroelectric phases transition in compounds, ferroelectric behavior in natural superlattice-structure, and strong magnetoelectric coupling in pillar–matrix multiferroic nanostructures.<sup>6-8</sup> However, the strength of magnetoelectric coupling is so far low that it is time to design and fabricate materials, it

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