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#### ACCEPTED MANUSCRIPT

# A Flexible Multiferroic Composite with High Self-biased Magnetoelectric Coupling

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#### **Abstract**

Polymer-matrix multiferroic composites with self-biased magnetoelectric (ME) coupling hold promises in flexible energy harvester, magnetic field sensors and actuators, etc. Using CoFe<sub>2</sub>O<sub>4</sub> (CFO) nanoparticles and multi-walled carbon nanotubes (CNTs) as fillers to the polyvinylidene fluoride (PVDF) matrix, piezomagnetic films (CFO-CNT-PVDF, also named as M layers) with different volume fractions of CFO and a fixed percentage of CNTs are obtained, which also electrode. By employing the layer of poly(vinylidene serve the fluoride-trifluoroethylene) (P(VDF-TrFE), named as P layer) sandwiched between the two conductive layers of CFO-CNT-PVDF, and followed by hot-pressing, three-layered multiferroic composite CFO-CNT-PVDF/P(VDF-TrFE)/ CFO-CNT-PVDF (named as M/P/M) are prepared. The ME coupling increases with the volume fraction of CFO nanoparticles. The maximum self-biased and peak values of ME coefficient ( $\alpha_{\text{ME}}$ ) reach 16.7 mV·cm<sup>-1</sup>·Oe<sup>-1</sup> and 25.8 mV·cm<sup>-1</sup>·Oe<sup>-1</sup>, respectively. Measurements on the relationship between the magnetostriction and static magnetic field  $(H_s)$  are performed for the CFO-CNT-PVDF films with different volume fraction of CFO. Based on the equivalent circuit model, the self-biased and peak values of  $\alpha_{\rm ME}$ 

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