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Spin-phonon coupling and ferroelectricity in magnetoelectric gallium ferrite[1]

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Gallium ferrite ($GaFeO_3$ or GFO) is a low temperature ferrimagnet and room temperature piezoelectric wherein the magnetic transition temperature (T_C) could be tailored to room temperature and above by tuning the stoichiometry and processing conditions. Such tunability of the magnetic transition temperature renders GFO a unique perspective in the research of multiferroics to potentially demonstrate room temperature magnetoelectric effect attractive for futuristic digital memory applications. Recent studies in several transition metal oxides highlight the importance of spin-phonon coupling in designing novel multiferroics by means of strain induced phase transition. In the present work, we have systematically studied the evolution of phonons in good quality samples of GFO across the T_C using Raman spectroscopy. Using the phonon softening behavior and nearest neighbor spin-spin correlation function below T_C we estimated spin-phonon coupling strength in the magnetically ordered state. In the process, we also show, for the first time, the presence of a spin glass phase in GFO where the spin-glass transition has a signature of abrupt change in spin-phonon coupling strength. Though GFO is piezoelectric and crystallizes in polar $Pc2_1n$ symmetry, its ferroelectric nature remained controversial probably due to the large leakage current in the bulk material. To address this issue, we deposited epitaxial thin film on single crystalline yttria stabilized zirconia (YSZ) substrate using indium tin oxide (ITO) as a bottom conducting layer. We demonstrate clear evidence of room temperature ferroelectricity in the thin films from the 180^o phase shift of the piezoresponse upon switching the electric field. Further, suppression of dielectric anomaly in presence of an external magnetic field clearly reveals a pronounced magneto-dielectric coupling across the magnetic transition temperature. In addition, using first principles calculations we elucidate that Fe ions are not only responsible for ferrimagnetism as observed earlier, but give rise to the observed ferroelectricity also, making GFO an unique single phase multiferroic.

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