Ferroelectric and Magnetic Properties in Room-Temperature Multiferroic $GaFeO_3$-type Thin Films

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Multiferroic materials exhibiting ferromagnetic and ferroelectric properties in a single phase have been eagerly studied due to their fascinating physics and novel technological applications such as fast-writing, power-saving, and non-destructive data storage. However, such multiferroic materials rarely exhibit both spontaneous magnetization and polarization at room temperature. $GaFeO_3$-type iron oxides are promising multiferroic materials due to the coexistence of a large spontaneous magnetization and polarization near and above room temperature as well as their multiferroic properties such as magnetic-field-induced modulation of polarization. GFO consists of one tetrahedral (T_d) Ga1 site and three octahedral (O_h) Ga2, Fe1, and Fe2 sites. The ferrimagnetism in GFO is derived from superexchange antiferromagnetic interactions between Fe ions, where the Fe ion magnetic moments at Ga1 and Fe1 sites are antiparallel to those at Ga2 and Fe2 sites. Especially, the Fe $3d^5$ orbital at the Fe2 site has strong hybridization with O $2p$ orbitals, producing a large spin-orbit interaction. This interaction leads to a much larger coercive field (H_c) of 20 kOe at 300 K in GFO-type Fe_2O_3 nanoparticles compared to other room-temperature multiferroic materials. For the ferroelectricity in GFO films, a unique mechanism of polarization switching is predicted. The displacement length of the metal cations at the Fe1 and Fe2 sites can reach as high as 1.2 Å during polarization switching, whereas that of conventional perovskite ferroelectrics such as $BaTiO_3$ is about 0.1 Å. Such a large displacement of magnetic Fe ions should realize a novel magnetoelectric effect.

To realize large magnetoelectric properties and applications of GFO films, the ferroelectric and ferrimagnetic properties at room temperature must be controlled. A key point for the existence of both ferroelectricity and ferrimagnetism at room temperature is to understand the relationship between the constituent composition at each cation site and the original character. Thus, a systematic investigation of multiferroicity as a function of the compositional ratio of Ga and Fe is important for a fundamental understanding and future applications. In this study, we fabricate high-quality $Ga_xFe_{2-x}O_3$ epitaxial thin films (x = 0.0–1.0) and systematically investigate their ferroelectric and ferrimagnetic properties. All films exhibit out-of-plane ferroelectricity and in-plane ferrimagnetism simultaneously. The coercive electric field (E_c) monotonically decreases with x. Additionally, increasing x reduces the coercive force (H_c) but enhances the saturated magnetization (M_s) at room temperature, according to the site of Ga ions. Finally, we demonstrate the room-temperature magnetocapacitance effects of the GFO films. The E_c, H_c, and M_s values can be widely controlled in ranges of 400–800 kV/cm, 1–8 kOe, and 0.2–0.6 μ_B/f.u at room temperature by changing x, respectively. Because such ferroelectric and magnetic ranges differ from those of well-known room-temperature multiferroic $BiFeO_3$, GFO-type iron oxides may expand the variety of room-temperature multiferroic materials.

Reference

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