

Growth of Ruddlesden–Popper $\text{Ca}_3\text{Mn}_2\text{O}_7$ thin films by Pulsed-Laser Deposition

Friday 24 September 2021 11:15 (1 minute)

Naturally Layered Perovskite structures with improper ferroelectricity [1, 2], such as the Ruddlesden-Popper calcium manganite compound $\text{Ca}_3\text{Mn}_2\text{O}_7$, offer an alternative route to achieve non-expensive and high-performance room temperature multiferroic magnetoelectricity for information storage, sensors, and actuators or low power energy-efficient electronics. They allow exploring oxygen octahedra nonpolar rotations and cation site displacement to attain non-centrosymmetry. Additionally, due to their high sensitivity to lattice-distortions, their preparation in thin film form over crystalline substrates allows the manipulation of acentricity and enables the tuning of lattice, electric and magnetic interactions. However, the preparation conditions to obtain the $\text{Ca}_3\text{Mn}_2\text{O}_7$ phase with the Ruddlesden-Popper structure need to be optimized and their properties have not yet been explored. As such, thin films of $\text{Ca}_3\text{Mn}_2\text{O}_7$ have been prepared over SrTiO_3 substrates by Pulsed Laser Deposition, using a $\text{Ca}_3\text{Mn}_2\text{O}_7$ target. Polycrystalline $\text{Ca}_3\text{Mn}_2\text{O}_7$ was synthesized using a conventional high-temperature ceramic route. The structural studies show that in the films prepared on SrTiO_3 , at 730 °C, with 4 J/cm² laser fluence, 10⁻³ mbar oxygen pressure and with a post-annealing process, the $\text{Ca}_2\text{Mn}_3\text{O}_7$ phase is stabilized, as confirmed by XRD and Raman Spectroscopy. The corresponding EDS analysis further gives a Ca/Mn atomic ratio of ~1.5:1, consistent with the presence of this phase. The magnetic properties were measured using a SQUID magnetometer, showing an antiferromagnetic transition at 110 K. The dielectric properties of the films show a relaxor-type behavior. The Havriliak-Negami function was fitted to the real and imaginary permittivity as a function of frequency (Fig. 1). The dielectric properties of the films will be discussed and presented, highlighting the phase evolution and stabilization in the films.

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Session Classification: Advanced Materials and Processes for Energy (Posters)