

Stabilization of an Extended Skyrmion Phase in Cu_2OSeO_3 via Finite-Size Effects

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We report a novel, facile, and comparatively low-power-consuming diffusion gel technique for the synthesis of a single-phase multiferroic Cu_2OSeO_3 in the bulk form. Initially, single crystals of chalcocite ($\text{CuSeO}_3 \cdot 2\text{H}_2\text{O}$) were grown via the gel method and subsequently converted to Cu_2OSeO_3 through controlled thermal decomposition. A detailed thermogravimetric analysis, combined with systematic experimentation under various annealing conditions such as annealing temperature, heating rate, and holding time, led to the optimization of single-phase Cu_2OSeO_3 formation from $\text{CuSeO}_3 \cdot 2\text{H}_2\text{O}$. The optimal annealing temperature was determined to be 475 ± 5 °C for a period of 24 hours. Structural characterization through XRD, FESEM, and TEM confirmed the formation of phase-pure Cu_2OSeO_3 with an average crystallite size (d) of 180 ± 10 nm, which is approximately three times larger than the helical spin modulation length ($\lambda \approx 60$ nm) reported for the magnetic skyrmion host Cu_2OSeO_3 . Temperature-dependent ac susceptibility and dc magnetization measurements, revealed a magnetic transition temperature $T_C \approx 59.25 \pm 0.25$ K. Anomalies in the ac susceptibility (χ_{ac}) as a function of magnetic field (H), measured at fixed temperatures (T) in the range 49 K - 61 K, indicates the presence of a skyrmion phase within a broad temperature range $0.85 T_C \leq T \leq 0.97 T_C$, significantly wider than that observed in single crystal Cu_2OSeO_3 . While the exchange stiffness (A_{ex}) derived from thermal demagnetization remains comparable to that of single-crystal Cu_2OSeO_3 , the Dzyaloshinsky–Moriya interaction (DMI) strength (D), inferred from the helical to field-polarized phase transition line obtained from $\chi_{ac}(H)$ data, exhibits a notable enhancement relative to conventional single- or polycrystalline Cu_2OSeO_3 . This enhancement is attributed to finite-size effects as $d \sim 3\lambda$, which facilitate the stabilization of a robust skyrmion lattice phase within the magnetic field–temperature (H – T) phase space.

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