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Investigation on external stimuli engendered magnetic ordering in polycrystalline $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ quadruple perovskite

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Abstract

The consequences of high energy mechanical milling, microwave-assisted heating and rapid thermal cooling on magnetic ordering in polycrystalline $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ cubic perovskite have been investigated by means of X-ray powder diffractometry (300 K), dc magnetization in field – cooled and zero – field cooled modes ($H = 100$ Oe and 1000 Oe, $T = 5 - 300$ K) ($M - T$ curves) and $M - H$ loop characteristics ($T = 5$ K and 300 K, $H_{\text{max}} = 70$ kOe). The $M - T$ curves of unmilled and 16 h milled samples show pure antiferromagnetic and weak ferromagnetic ordering, respectively, 1 h and 6 h milled samples demonstrate the coexistence of both the phases while microwave-assisted and quenched samples exhibit classic antiferromagnetic transition and a low temperature paramagnetic-like contribution with different weights, well supported by the $M - H$ loop characteristics. The observed transformations in the magnetic ordering are attributed to the ball-milling induced stress which curtails hybridization of empty Ti-3d orbitals with Cu-3d and O-2p orbitals and secondary phase formation. Oxygen vacancies associated with bound magnetic polarons originate ferromagnetism in the milled samples while unpaired electrons inhabited at the empty sites are the cause of paramagnetic centers. The low-temperature Curie – tail in $M - T$ curve for quenched and microwave assisted samples is attributed to Ti^{3+} cations.

Keywords: $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$, magnetic ordering, mechanical milling, orbital hybridization, oxygen vacancies.

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