Signature of field-induced spin ice state and evolution of structural and magnetic phase on La substitution in disordered pyrochlore oxide $Dy_2Zr_2O_7$

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Dy₂Zr₂O₇ exhibits Dy₂Ti₂O₇ type high temperature magnetic field induced spin freezing near ~ 10 K in ac susceptibility measurements [1]. The magnetic heat capacity of Dy₂Zr₂O₇ shows a correlation peak at 2 K, but no residual entropy was observed. The low-temperature magnetic entropy at 5 kOe field is R[ln2 - 1/2ln(3/2)] which is the same as for the spin ice state. Substitution of non-magnetic, isovalent La³⁺ for Dy³⁺ gradually induces the structural change from highly disordered fluorite to stable pyrochlore phase through a biphasic mixture of both. We observed that the higher La compositions ($1.5 \le x \le 1.9$), show spin freezing (T ~ 17 K) similar to the field induced spin ice freezing for low La compositions ($0 \le x \le 0.5$), and the well-known spin ice systems Dy₂Ti₂O₇ and Ho₂Ti₂O₇. The low temperature magnetic state for higher La compositions ($1.5 \le x \le 1.9$) culminates into spin glass state below 6 K. The Cole-Cole plot and Casimir-du Pré fit shows narrow distribution of spin relaxation time in these compounds.

References:

[1] J. Snyder et al., Nature, 413, 48 (2001)