Magnetic Ground-State Engineering in Multiferroic Oxides for Cryogenic Cooling

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Hexagonal manganites (RMnO$_3$, R = Sc, Y, In, and Dy-Lu) are multifunctional materials exhibiting, for example, improper ferroelectricity, strong magnetoelectric coupling, and the thermal Hall effect. At cryogenic temperatures, the interplay between 4f and 3d magnetic moments leads to complex magnetic phases, ranging from long-range antiferromagnetism to frustrated spin states. Additional unusual responses arise from the coupling between magnetic and ferroelectric orders, because of which the RMnO$_3$ family became a model system for multiferroicity. More recently, high cooling efficiency at cryogenic temperatures was reported utilizing the magnetocaloric effect (MCE), i.e., reversible temperatures change induced by a magnetic field, making RMnO$_3$ systems interesting for refrigeration in quantum technologies and gas liquefaction.

Here, we investigate the cryogenic MCE in polycrystalline hexagonal manganites and isostructural oxides. In hexagonal manganites, direct adiabatic temperature measurements in pulsed magnetic fields reveal reversible temperature changes of up to 20 K at fields above 20 T (**Fig. 1**). The MCE scales directly with the effective magnetic moment on the R-site. We compare these results to RInO$_3$ compounds, where the absence of magnetic 3d moments can induce frustration of magnetic order on the R-site. For DyInO$_3$, magnetic-field-dependent susceptibility and specific-heat measurements confirm spin-liquid-like behavior down to the milliKelvin regime. Owing to its magnetic frustration, DyInO$_3$ exhibits a pronounced MCE that peaks near 2 K, significantly lower than in the hexagonal manganites, while adiabatic demagnetization experiments reveal cooling to temperatures below 240 mK.

Our results establish polycrystalline hexagonal oxides as a chemically tunable platform for magnetic ground-state engineering and highlight their potential for cryogenic refrigeration technologies. In particular, the complex magnetism of these materials opens perspectives for energy-efficient electric-field-assisted control of cryogenic caloric responses.

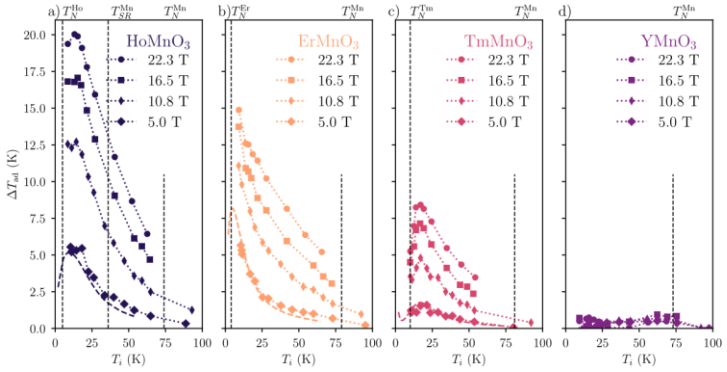

Figure 1: Adiabatic temperature changes under pulsed magnetic fields. Data are shown for (a) HoMnO$_3$, (b) ErMnO$_3$, (c) TmMnO$_3$, and (d) YMnO$_3$. Symbols represent directly measured adiabatic temperature changes as a function of the initial temperature, T_i, for magnetic-field pulses between 5.0 and 22.3 T. Dotted lines are guides to the eye. Dashed lines show values calculated from entropy data measured under 5 T, while dashed vertical lines indicate the magnetic ordering temperatures.

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