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Anomalous specific heat and magnetic properties of $Tm_xDy_{1-x}Al_2$ ($0 \leq x \leq 1$)Arjun K. Pathak,^{1,a} C. M. Bonilla,¹ D. Paudyal,¹ Y. Mudryk,¹ and V. K. Pecharsky^{1,2}¹*The Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011-3020, USA*²*Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011-2300, USA***Abstract:**

We study crystal structure, phase transitions and magnetism of pseudo-binary $Tm_xDy_{1-x}Al_2$ ($0 \leq x \leq 1$) compounds using temperature dependent X-ray powder diffraction, specific heat and magnetization measurements, first principles, and model calculations. In low external magnetic fields, Dy-rich compounds undergo continuous, second-order phase transitions at the respective Curie temperatures, T_C . In contrast, the Tm-rich compounds exhibit discontinuous, first-order anomalies in the magnetically ordered states. These sharp transitions correlate with a substantial energy difference between the room temperature cubic and ground state rhombohedral structures of $TmAl_2$. A clear anomaly in the lattice parameter is observed at ~ 30 K for $x = 0.5$, which nearly coincides with $T_C = 31.2$ K. The effective quadrupolar moment of the lanthanides changes sign around $x = 0.5$, which leads to a nearly zero anisotropy constant and approximately spherical effective $4f$ charge densities, providing an explanation for the lack of structural distortions below T_C for $x = 0.5$. The calculations confirm [001] as the easy magnetization axis in the ground state tetragonal structure of $DyAl_2$, and reveal collapse of the orbital magnetic

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