

## *Supplementary materials*

# Tuning Room-Temperature Ferromagnetism in High-Entropy Oxide Thin Films via Vacuum Annealing-Induced Rocksalt-to-Spinel Phase Transition

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Table S1. Surface elemental compositions of different samples (at.%)

Sample	O	Al	Cr	Mn	Fe	Ni
T-Original	58.6	8.7	7.2	7.5	8.9	9.1
T773	58.8	8.5	7.5	7.3	9.0	8.9
T973	57.9	8.5	7.6	7.7	9.3	9.0
T1173	57.4	8.6	7.7	7.9	8.9	9.5

**Note:** Table S1 presents the surface elemental compositions of different samples obtained from XPS analysis. As shown in the table, the various metal elements in the HEO films are mixed in near-equimolar proportions. Combined with the XRD and EDS-mapping data in the main text, these films can be identified as high-entropy oxides. On the other hand, regardless of whether they possess a rocksalt or spinel structure, the HEO films deposited in this study consistently exhibit an oxygen-rich and metal-deficient state. Such a defective oxide state provides the compositional prerequisite for the heat-treatment-induced phase transition from rocksalt to spinel.

It should be noted that from a stoichiometric perspective, while a perfect rock-salt to spinel transition requires external oxygen ( $3\text{MeO} + 0.5\text{O}_2 \rightarrow \text{Me}_3\text{O}_4$ ), our room-temperature deposited HEO films are highly defective. XPS data (Table S1) reveals an initial non-equilibrium state with metal deficiency and oxygen excess, which can be formulated as  $\text{Me}_{1-x}\text{O}$ . Since the overall oxygen content remains stable during vacuum annealing, the phase transition does not rely on external oxygen

absorption. Instead, driven by the lower Gibbs free energy of the spinel phase, it occurs purely via the internal migration and rearrangement of pre-existing cations and defects. This process transforms the metastable rock-salt ( $\text{Me}_{1-x}\text{O}$ ) into the ordered spinel ( $\text{Me}_{3-x}\text{O}_4$ ) structure, completely corroborating the phase transition observed in our XRD and TEM results.

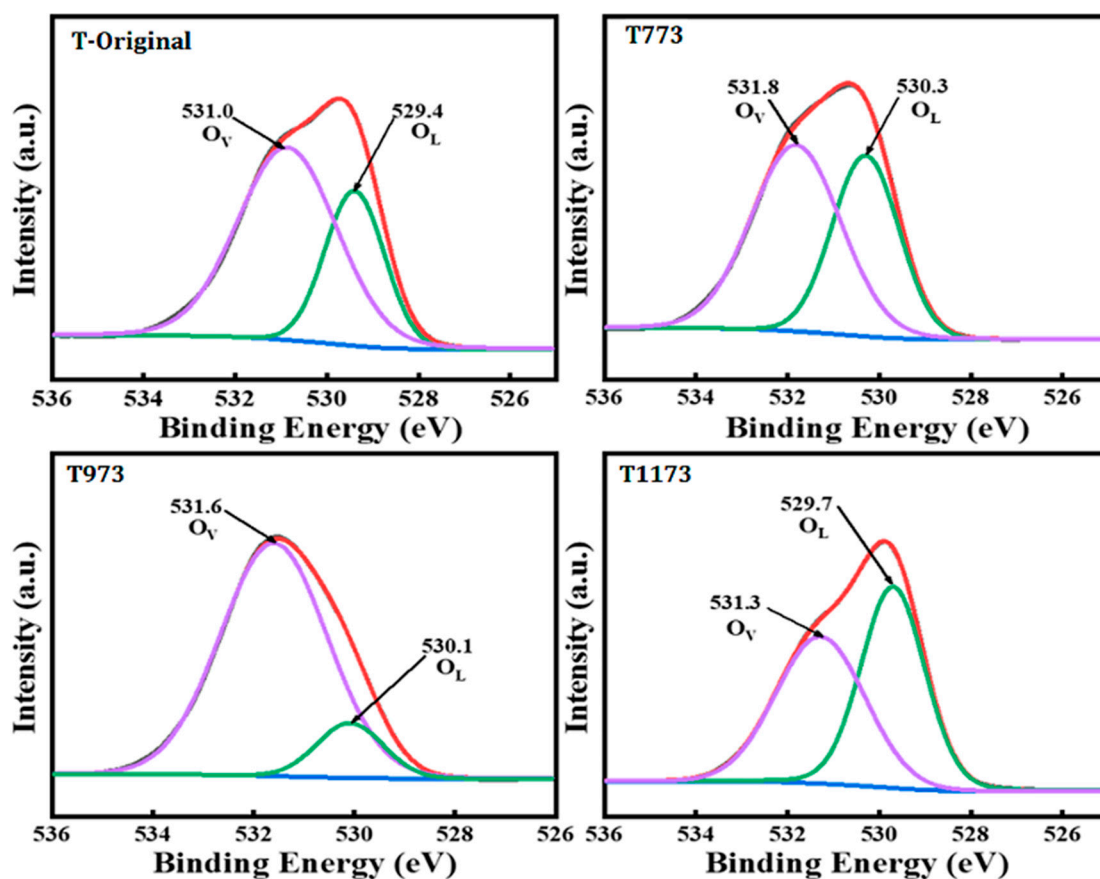


Figure S1. Deconvolution analysis of the O 1s peak in the high-resolution XPS spectra of the HEO thin films.

Table S2. Relative contents of lattice oxygen and oxygen vacancies in the films

Sample	O <sub>L</sub>	O <sub>V</sub>
T-Original	42%	58%
T773	36%	64%
T973	13%	87%
T1173	48%	52%

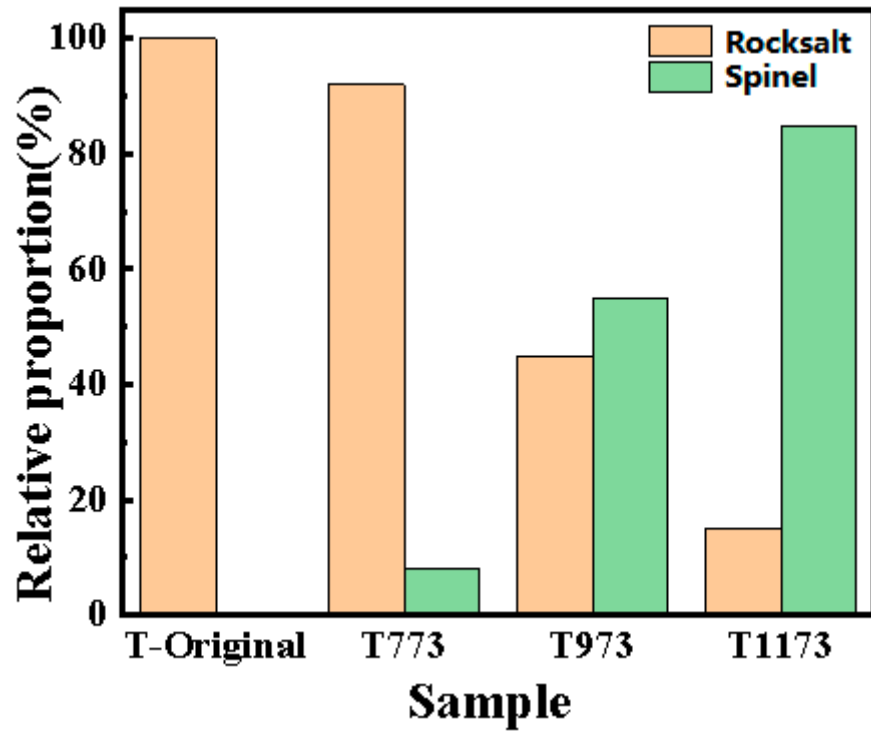


Figure S2. Phase content estimation (based on the RIR method using Jade 6.0)