Supplementary information for "Mg-doping enhanced magnetoelectric effect in polar magnet $Fe_2Mo_3O_8$"

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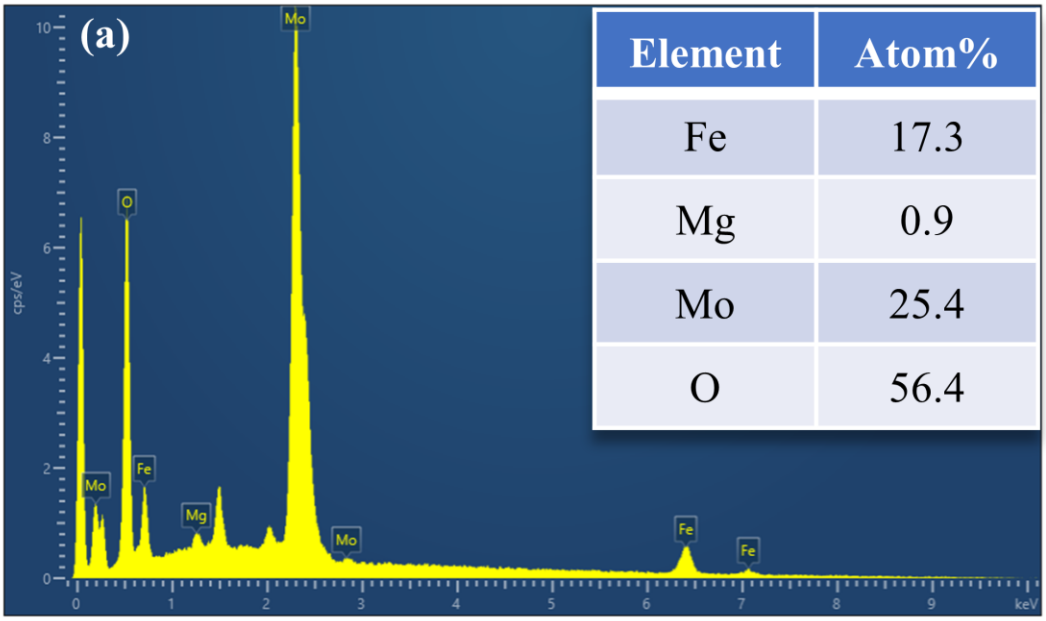

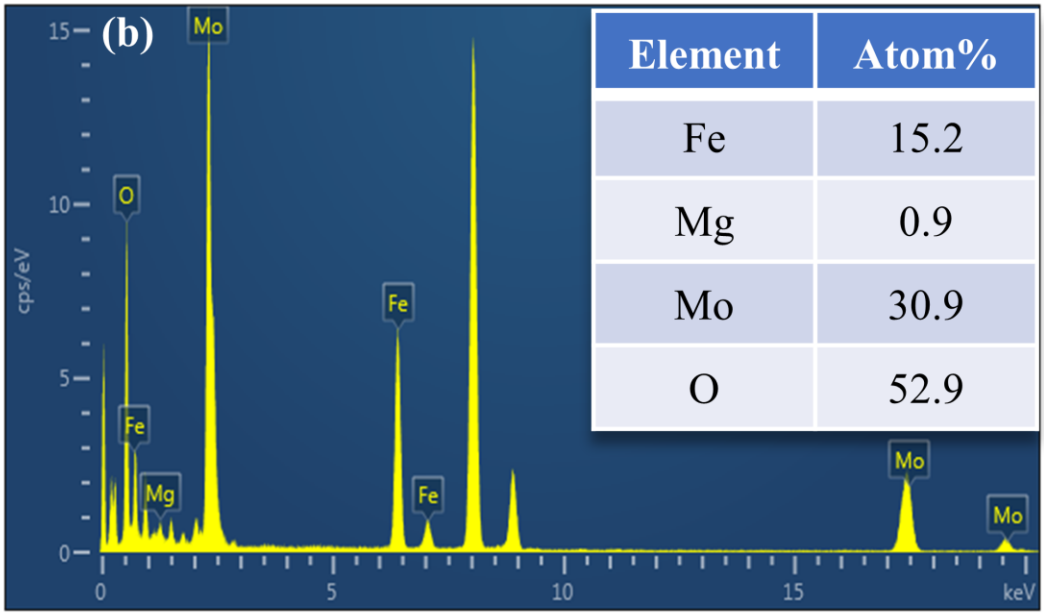

FIG. S1. Chemical compositions analysis of FMMO. (a) SEM-EDX and (b) TEM-EDX spectra with chemical compositions analysis of sample.

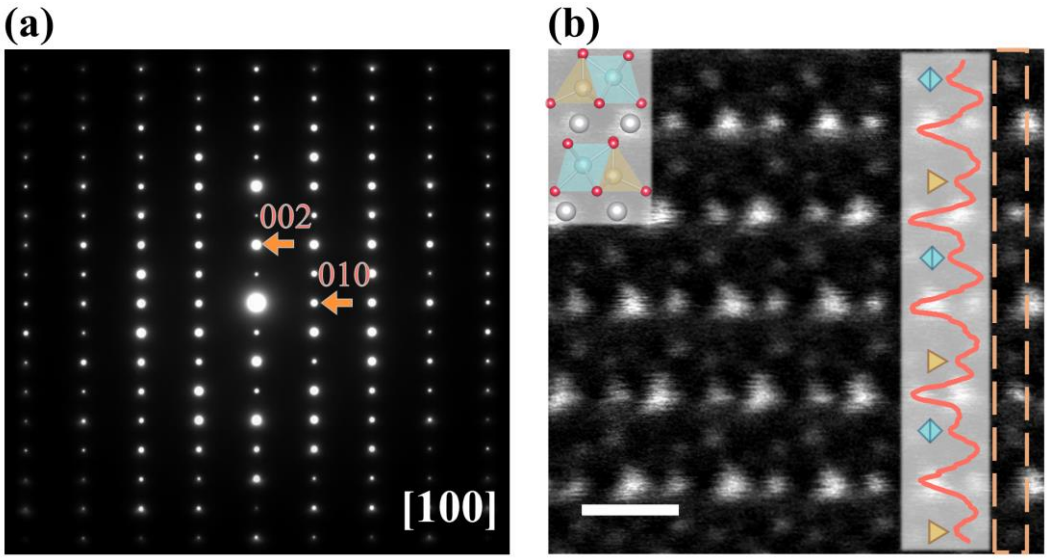

FIG. S2. Structural characterizations of $Fe_2Mo_3O_8$. (a) SAED pattern along the [100] zone axis. (b) Corresponding HAADF image with an overlaid structural model and scale bar being 0.5 nm. White and red spheres represent Mo and O atoms, respectively. Yellow and blue spheres denote Fe atoms in tetrahedrons and octahedrons, respectively. Red curve is the line profile corresponding to orange dashed box. Yellow and blue symbols refer to positions of tetrahedral and octahedral oxygen coordinations, respectively.

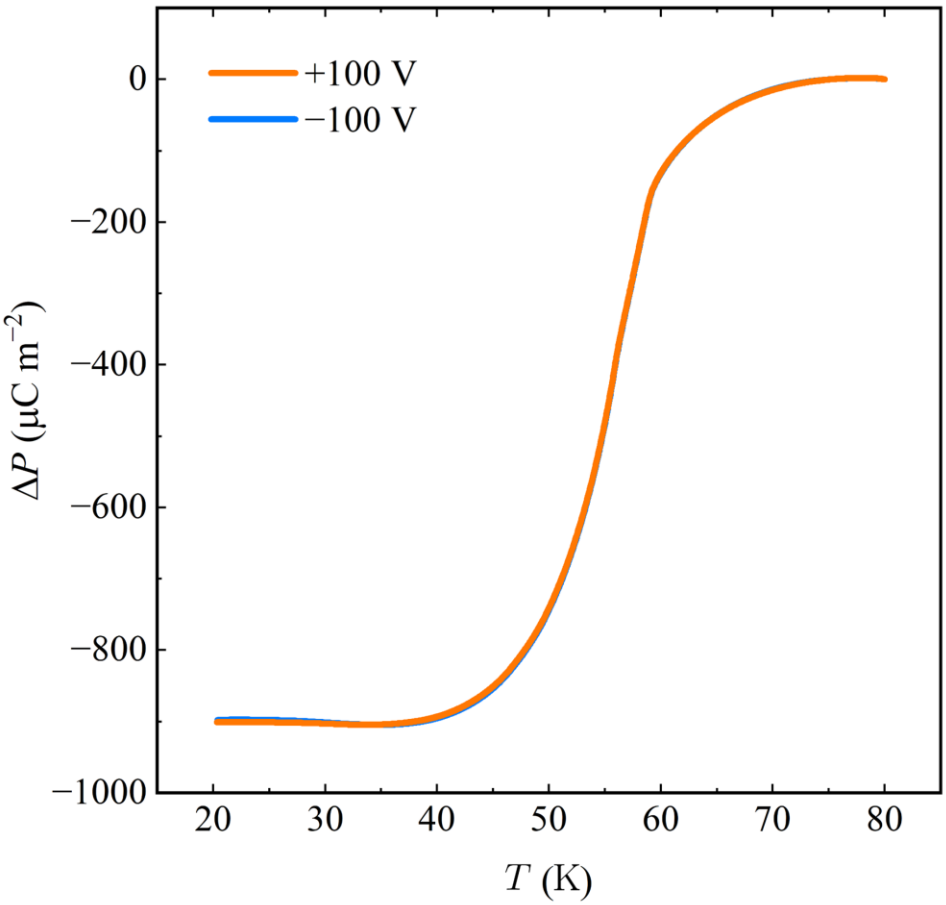

FIG. S3. Evidence of pyroelectricity in FMMO. Polarization changes along *c* axis as a function of temperature, showing that the polarization does not reverse its sign after a negative poling.

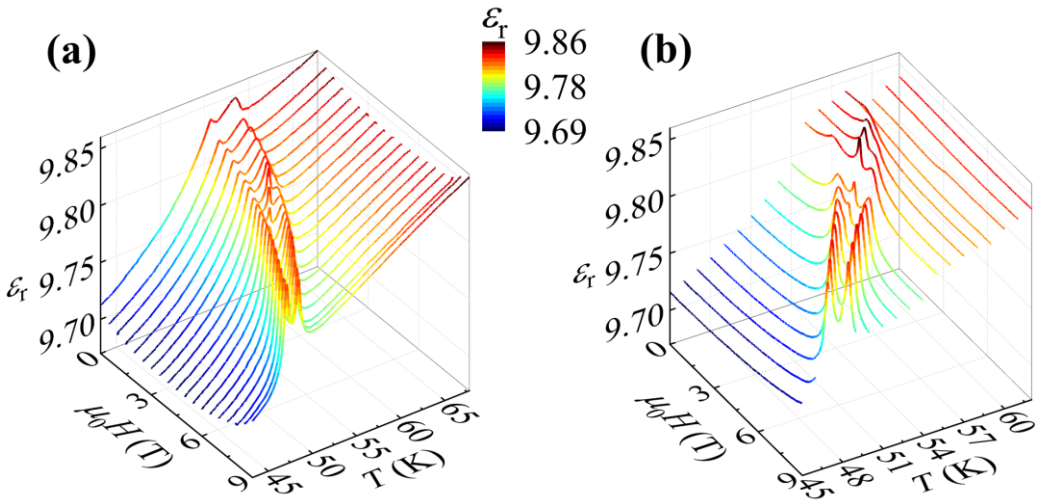

FIG. S4. Dielectric property of FMMO. (a) Temperature-dependent relative dielectric constant $\varepsilon_r(T)$ at various magnetic fields. (b) Magnetic-field-dependent relative dielectric constant $\varepsilon_r(H)$ at various temperatures. Color scale is shown in the middle.

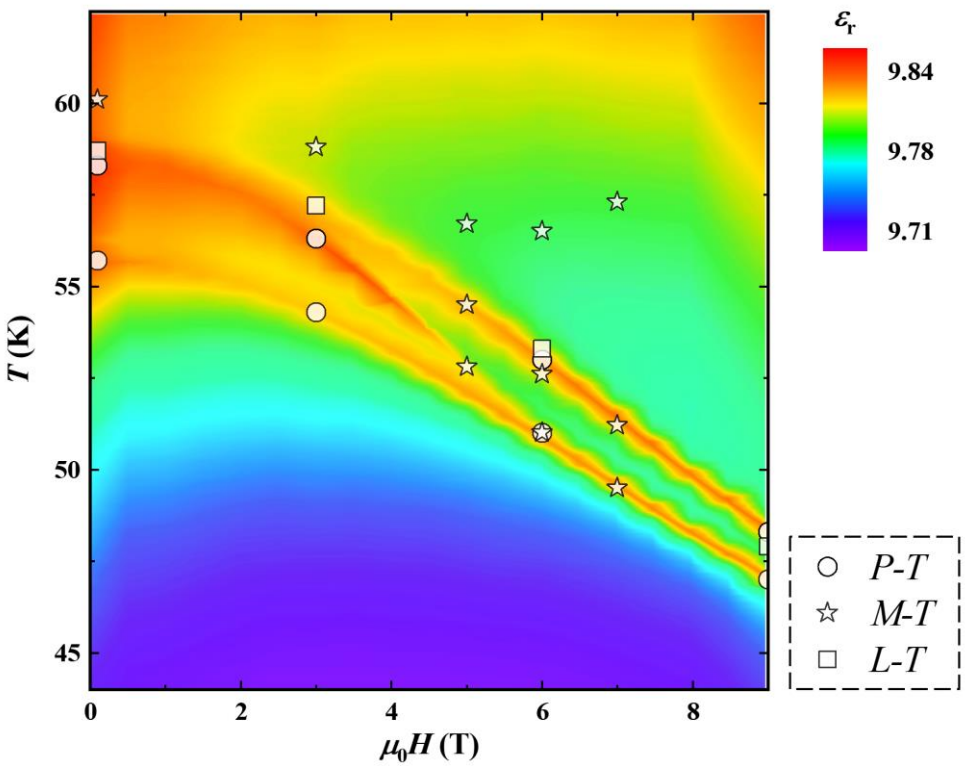

FIG. S5. *H-T* phase diagram with stepwise feature determined by *T*-scan. Phase diagram is determined by variations of relative dielectric constant ε_r (color scale at upper right). Empty circles, empty asterisks, and empty squares denote the positions of stepwise characteristic in $P(T)$, $M(T)$, and $L(T)$, respectively.