Supporting Information (Online Material) for

Hybrid Multiferroic Behavior in the Double Perovskite $(Ca_{0.5}Mn_{1.5})MnWO_6$

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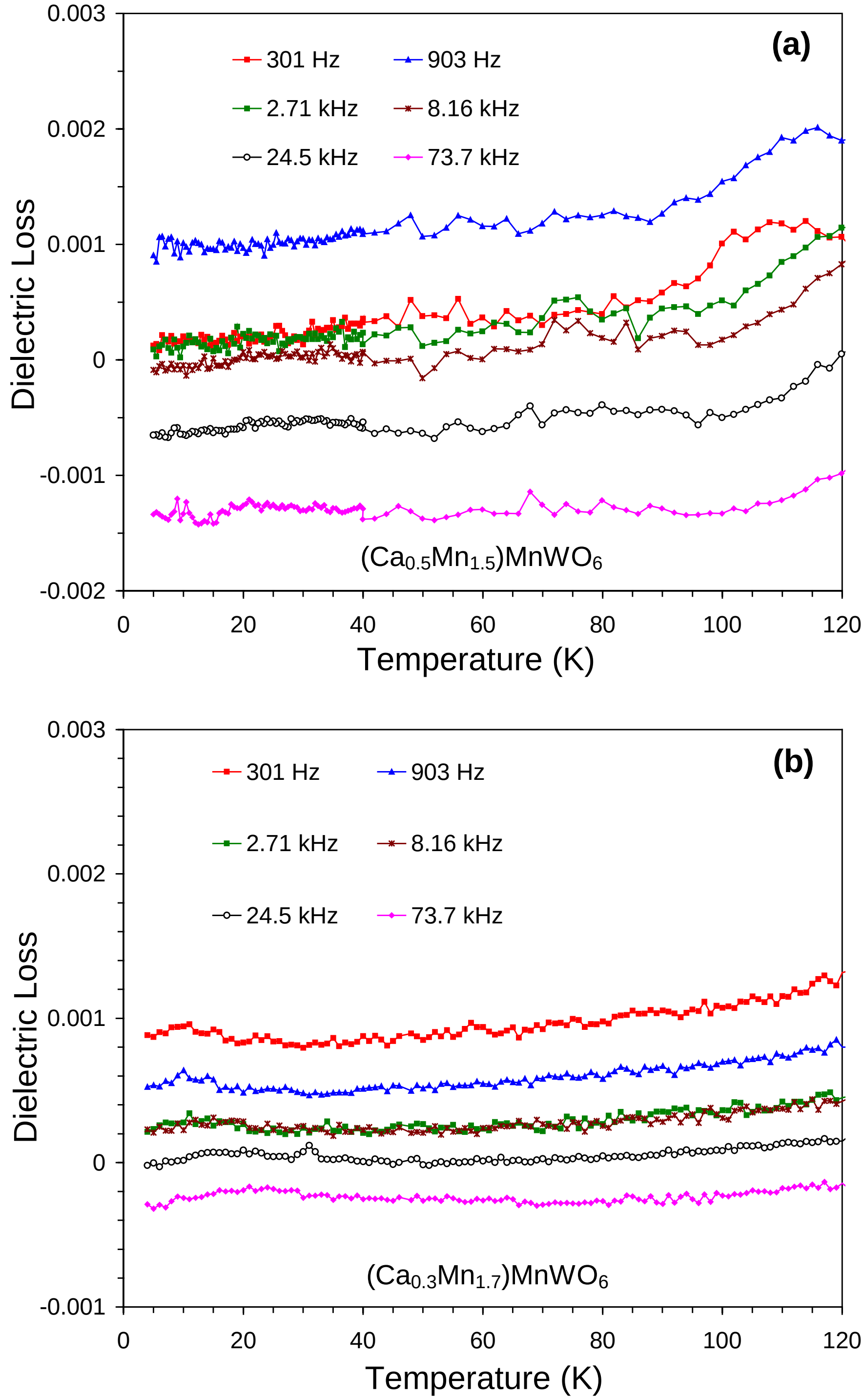


Figure S1. Temperature dependence of dielectric loss in (a) $(Ca_{0.5}Mn_{1.5})MnWO_6$ and (b) $(Ca_{0.3}Mn_{1.7})MnWO_6$ between T = 3 K and 120 K at different frequencies (f from 301 Hz to 73.7 kHz) at zero magnetic field.

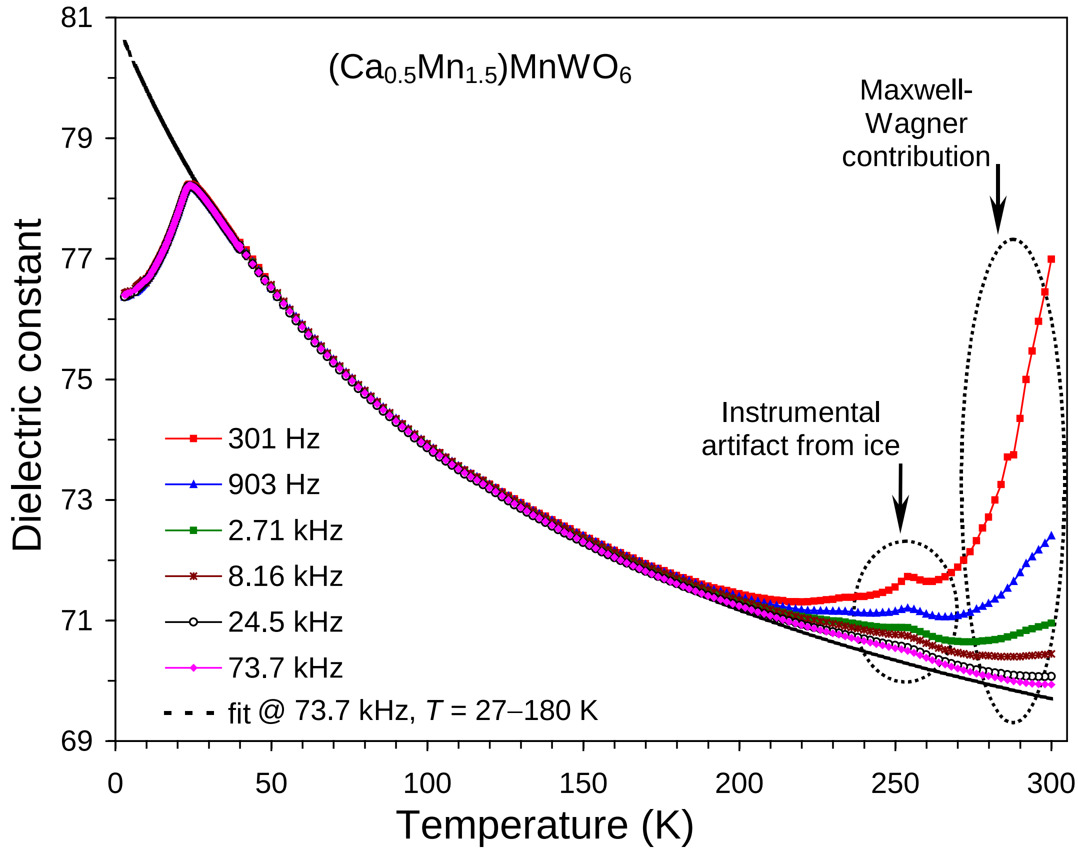


Figure S2. Temperature dependence of dielectric constant of $(Ca_{0.5}Mn_{1.5})MnWO_6$ between T = 3 K and 300 K at different frequencies (f from 301 Hz to 73.7 kHz) at zero magnetic field. The black line shows a fit (between 27 K and 180 K using data at f = 73.7 kHz) by the Curie-Weiss law, where the calculated curve was extended down to 3 K and up to 300 K using the obtained fitted parameters. Anomalies near 250 K (highlighted by a broken oval) are instrumental artifacts originating from ice as such anomalies were observed in many other samples measured on the same instrument (PPMS). Maxwell-Wagner contributions at low frequencies are highlighted by a broken oval.

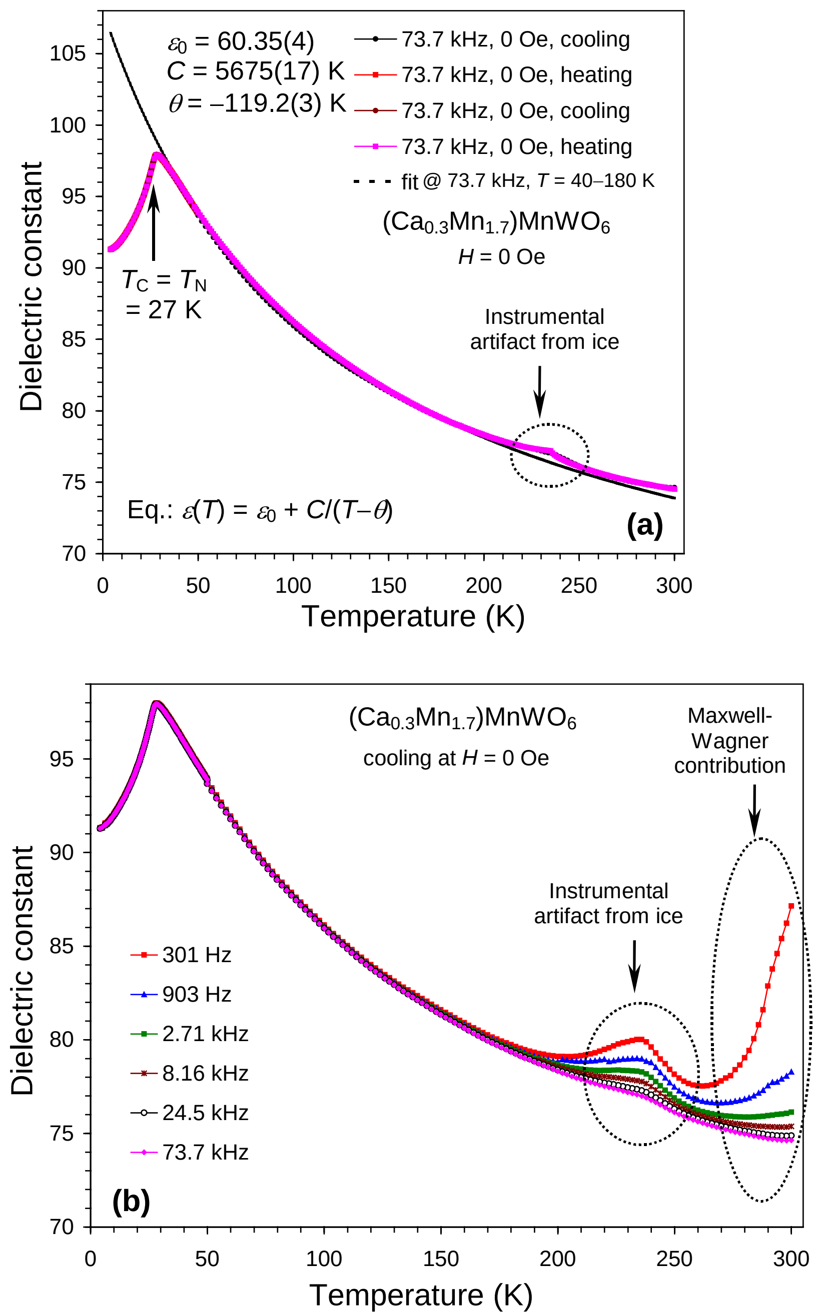
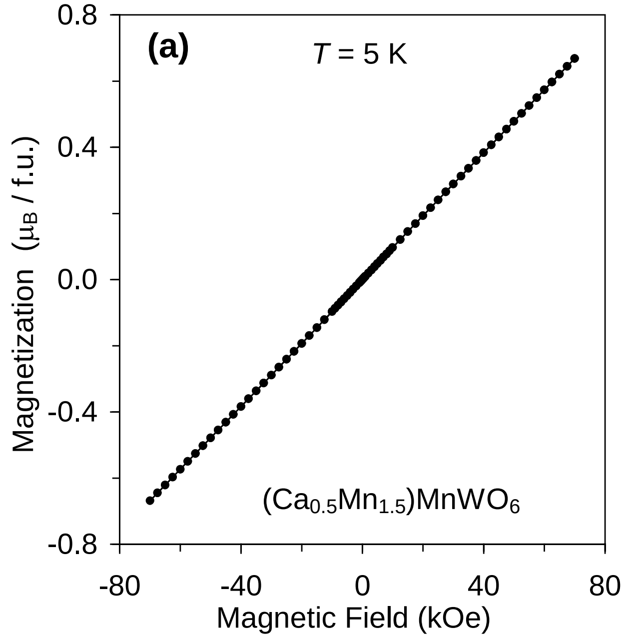


Figure S3. (a) Temperature dependence of dielectric constant of $(Ca_{0.3}Mn_{1.7})MnWO_6$ between T = 3 K and 300 K at one frequency of 73.7 kHz at zero magnetic field on cooling and heating (two runs). The black line shows a fit (between 40 K and 180 K) by the Curie-Weiss law, where the calculated curve was extended down to 3 K and up to 300 K using the obtained fitted parameters. Anomalies near 250 K (highlighted by a broken oval) are instrumental artifacts originating from ice. (b) Temperature dependence of dielectric constant of $(Ca_{0.3}Mn_{1.7})MnWO_6$ between T = 3 K and 300 K at different frequencies (f from 301 Hz to 73.7 kHz) at zero magnetic field. Maxwell-Wagner contributions at low frequencies are highlighted by a broken oval.



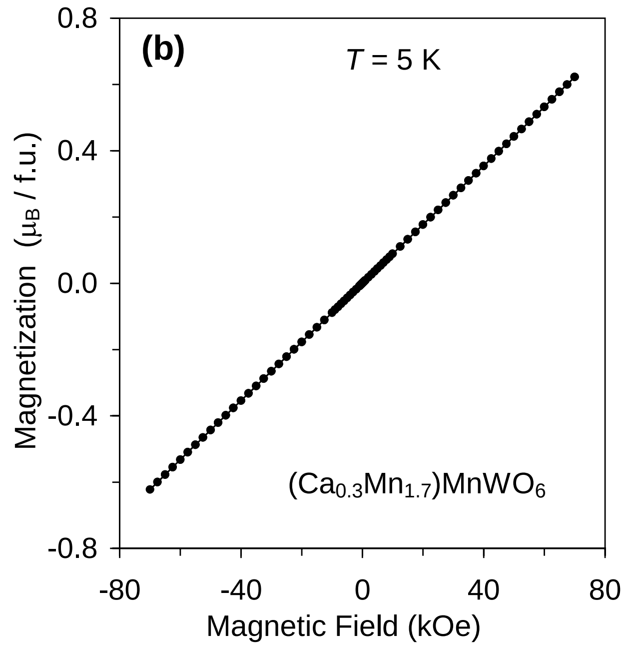


Figure S4. (a) An *M* versus *H* curve of $(Ca_{0.5}Mn_{1.5})MnWO_6$ at T = 5 K. (b) An *M* versus *H* curve of $(Ca_{0.3}Mn_{1.7})MnWO_6$ at T = 5 K.

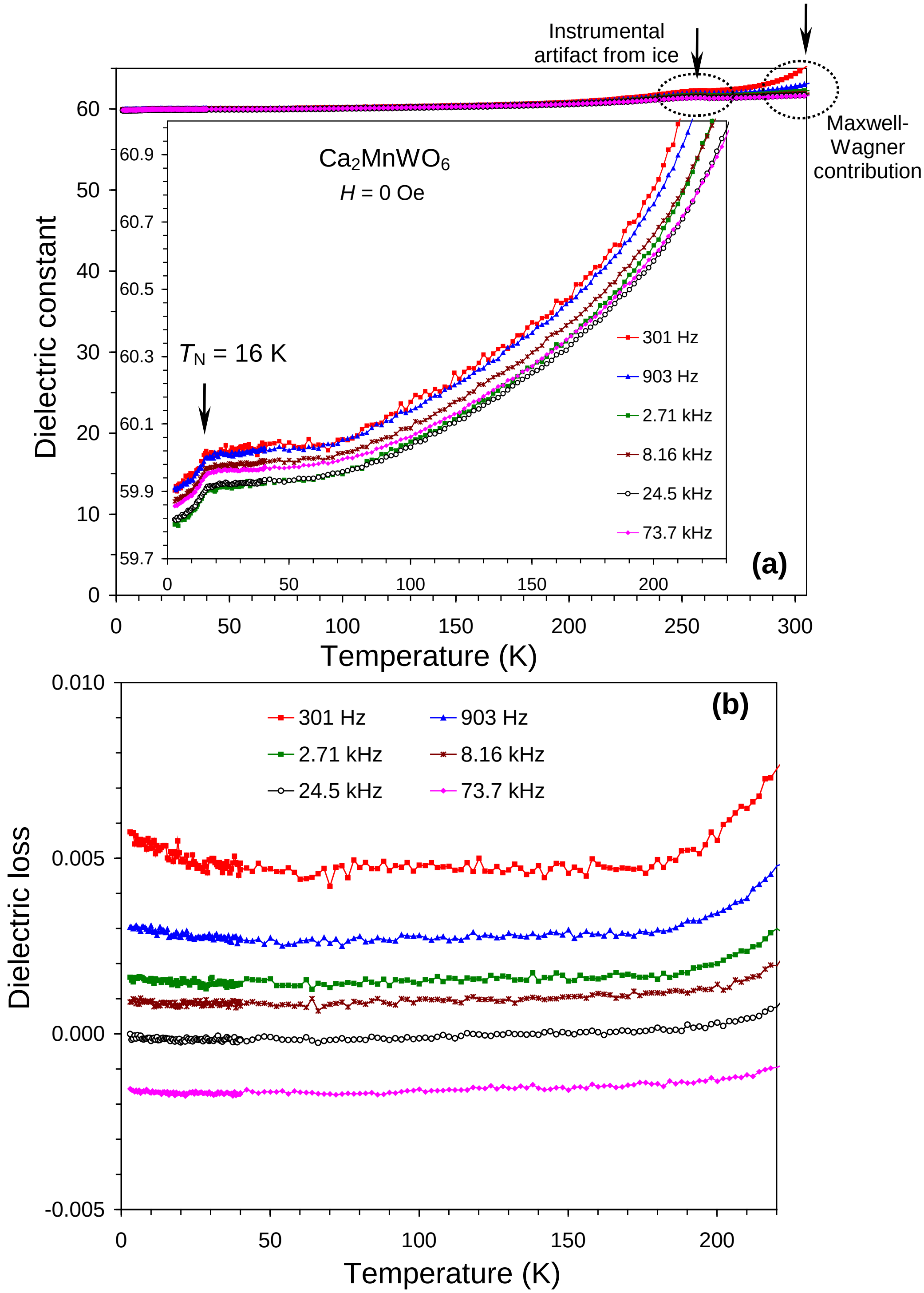


Figure S5. (a) Temperature dependence of dielectric constant of Ca_2MnWO_6 between T = 3 K and 300 K at different frequencies at zero magnetic field. Anomalies near 250 K (highlighted by a broken oval) are instrumental artifacts originating from ice. Maxwell-Wagner contributions at low frequencies are highlighted by a broken oval. Inset shows details between 3 K and 230 K. (b) Temperature dependence of dielectric loss of Ca_2MnWO_6 below 220 K.

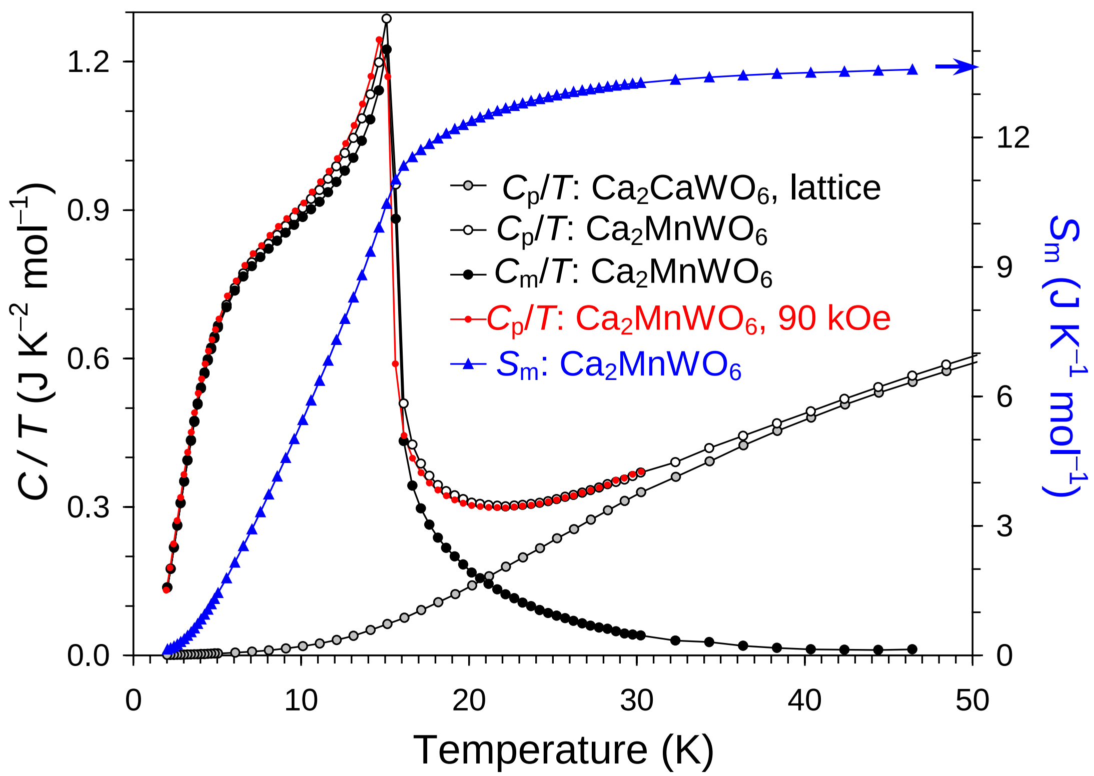


Figure S6. Specific heat data for Ca_2MnWO_6. White circles: C_p/T versus T at H = 0 Oe; black circles: C_m/T versus T at H = 0 Oe (where C_m is the magnetic part of specific heat); blue triangles and the right-hand axis: S_m versus T at H = 0 Oe (where S_m is magnetic entropy); red small circles: C_p/T versus T at H = 90 kOe. Gray circles show C_p/T versus T data at H = 0 Oe for Ca_2CaWO_6, which can serve as a lattice estimation. All data were measured on cooling. The experimental magnetic entropy of 13.57 J K^{-1} mol^{-1} was close to the expected value of 14.90 J K^{-1} mol^{-1} for Mn^{2+}.

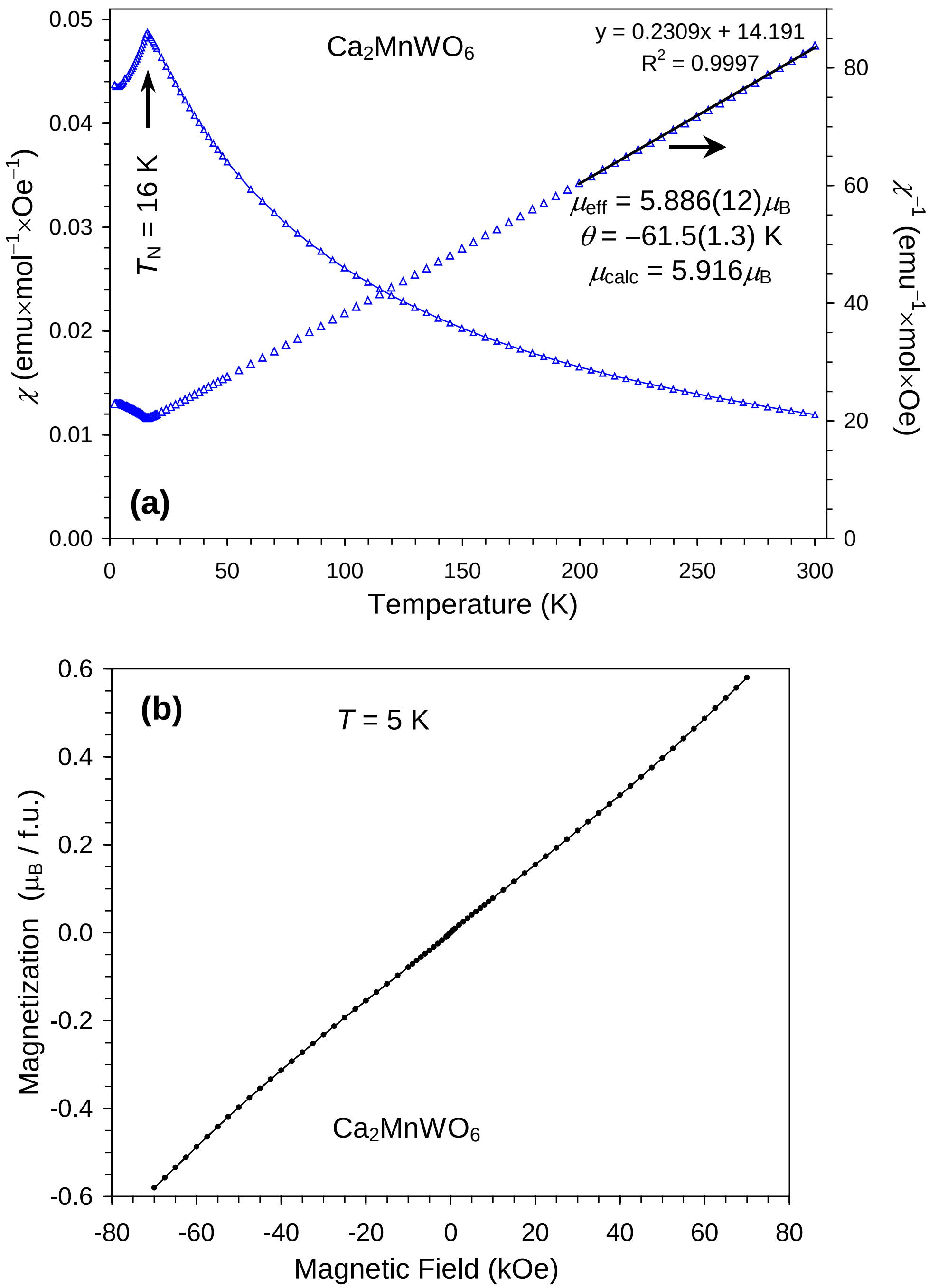


Figure S7. (a) A field-cooled on cooling (FCC) dc magnetic susceptibility ($\chi = M/H$) curve of Ca_2MnWO_6 measured at H = 10 kOe. Right-hand axis shows the χ^{-1} versus T curve with the Curie-Weiss fit (black line). Parameters of the fit are shown on the figure. (b) An M versus H curve of Ca_2MnWO_6 at T = 5 K.

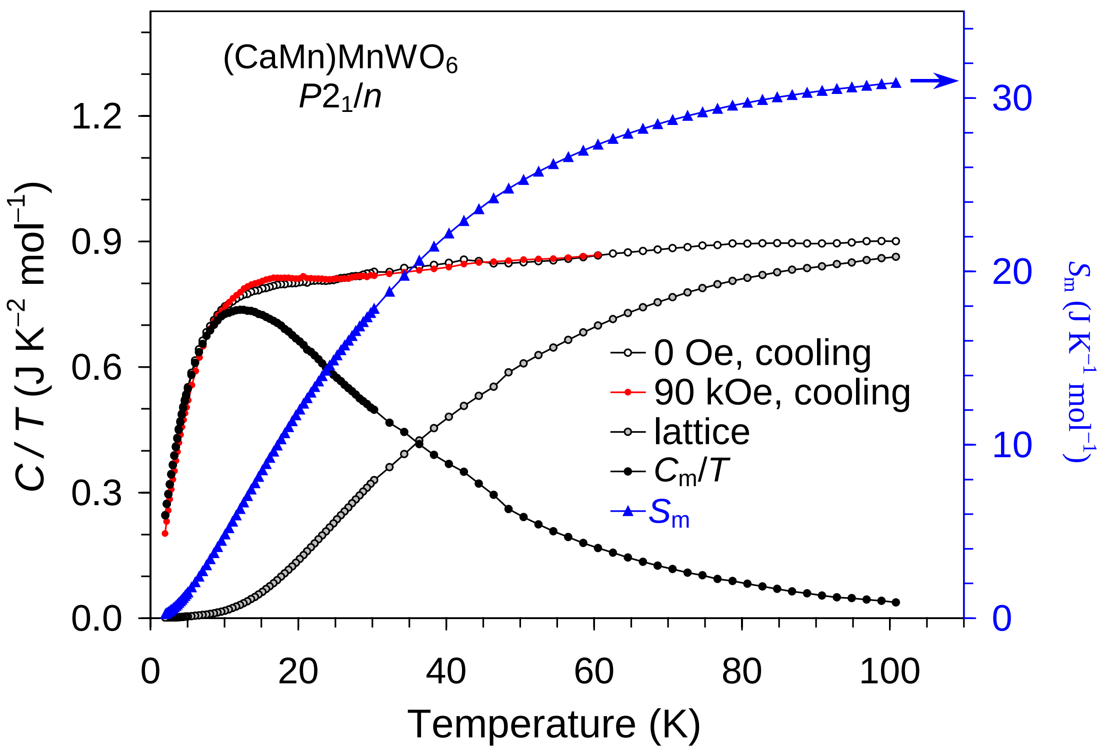


Figure S8. Specific heat data for (CaMn)$MnWO_6$ ($P2_1/n$ modification). White circles: C_p/T versus T at H = 0 Oe; black circles: C_m/T versus T at H = 0 Oe (where C_m is the magnetic part of specific heat); blue triangles and the right-hand axis: S_m versus T at H = 0 Oe (where S_m is magnetic entropy); red small circles: C_p/T versus T at H = 90 kOe. Gray circles show a lattice contribution (C_{lat}/T versus T) at H = 0 Oe, where C_{lat} is C_p for Ca_2CaWO_6 between 2 K and 46 K and C_p for Ca_2MnWO_6 between 48 K and 100 K. All data were measured on cooling. The experimental magnetic entropy of about 30.9 J K^{-1} mol^{-1} was close to the expected value of 29.8 J K^{-1} mol^{-1} for $2Mn^{2+}$. A broad anomaly is only seen due to short-range magnetic ordering. A very weak anomaly near 45 K is from a ferrimagnetic long-range ordering in the $P4_2/n$ modification.

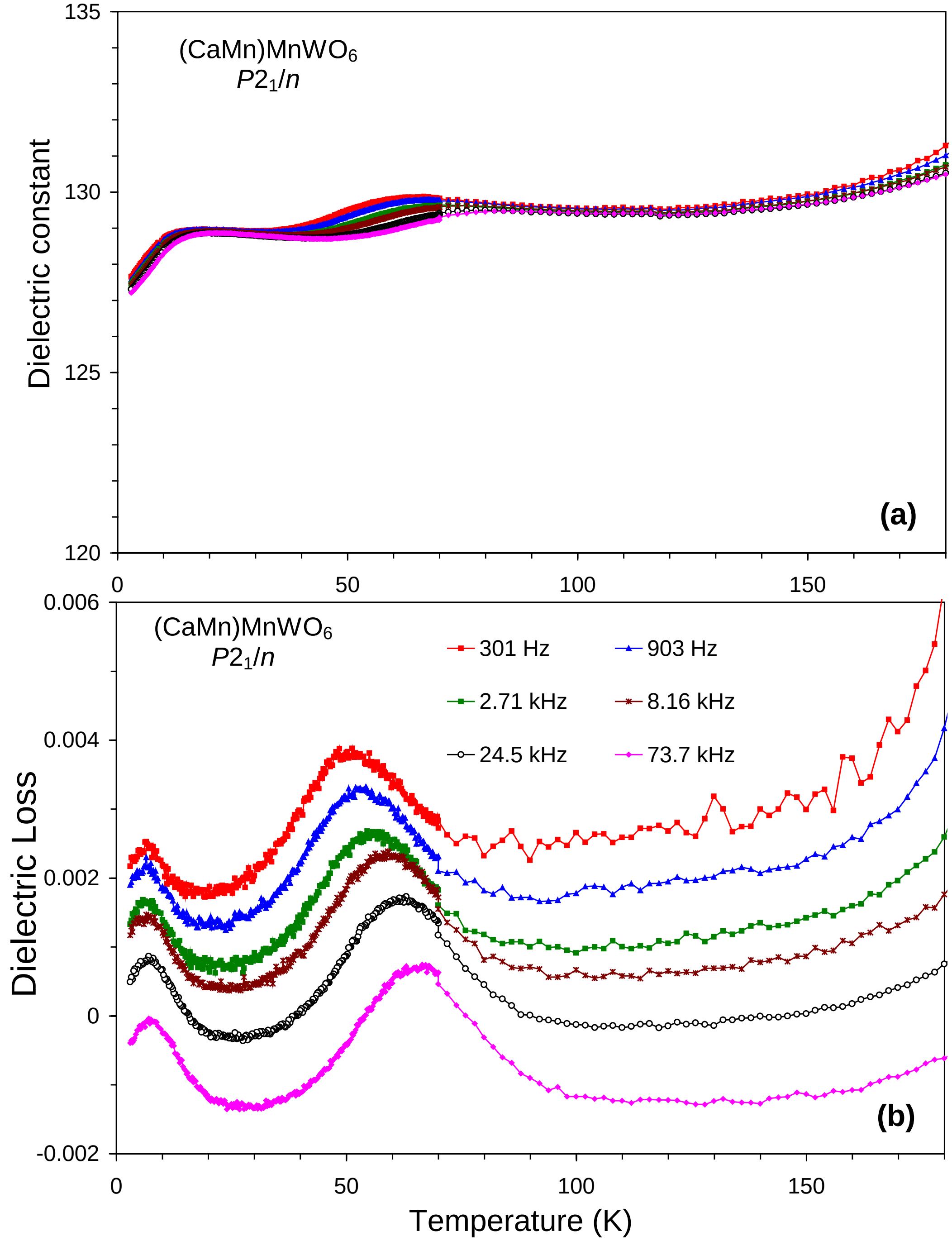


Figure S9. (a) Temperature dependence of dielectric constant of (CaMn)$MnWO_6$ ($P2_1/n$ modification) between T = 3 K and 180 K at different frequencies at zero magnetic field. (b) Temperature dependence of dielectric loss of (CaMn)$MnWO_6$. Relaxation-like behavior clearly seen near 50 K in the shifts of dielectric loss peaks as a function of frequency can be explained by a small admixture of the $P4_2/n$ modification and interfacial effects. Note that the $P4_2/n$ modification has a long-range ferrimagnetic transition at 45 K.