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Supplementary Materials for

Giant positive magnetoresistance in half-metallic double-perovskite Sr_2CrWO_6 thin films

Ji Zhang, Wei-Jing Ji, Jie Xu, Xiao-Yu Geng, Jian Zhou, Zheng-Bin Gu, Shu-Hua Yao, Shan-Tao Zhang

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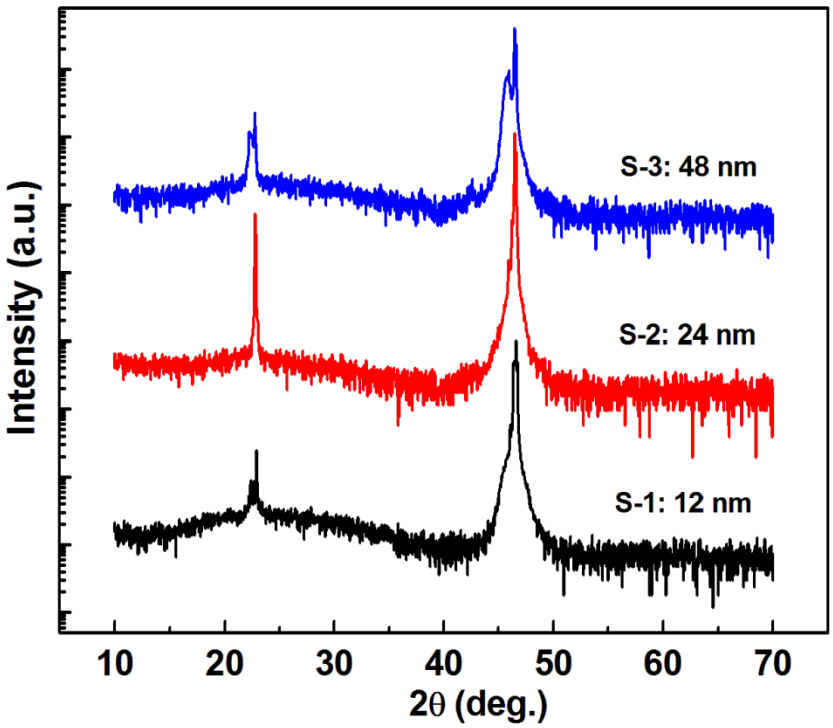

fig. S1. XRD patterns of the SCWO films with different thicknesses. The thickness of S-1, S-2 and S-3 is 12 nm, 24 nm and 48 nm respectively.

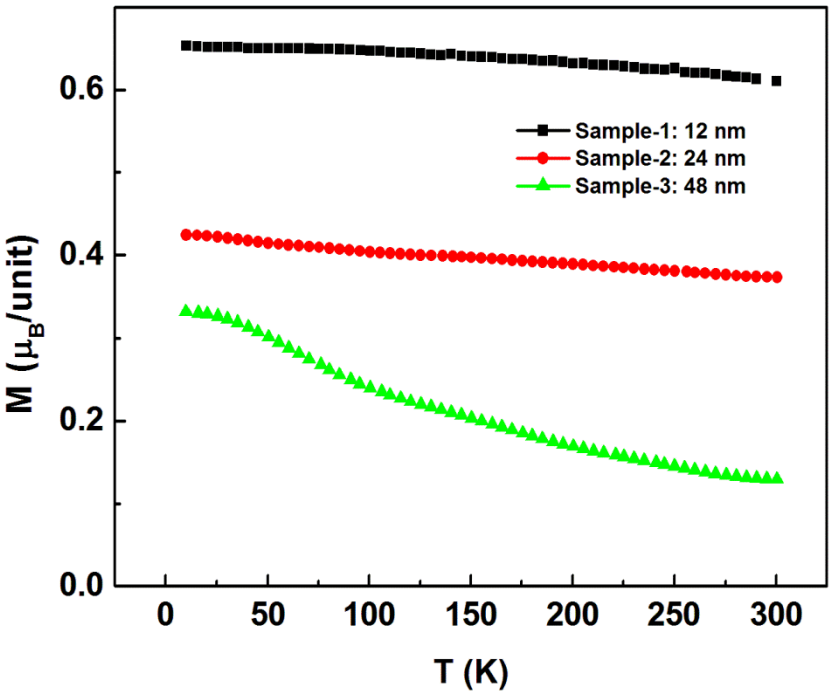

fig. S2. Field cooling (200 Oe) temperature-dependent magnetization of the S-1, S-2, and S-3 films. The results confirm the ferrimagnetic nature of SCWO.

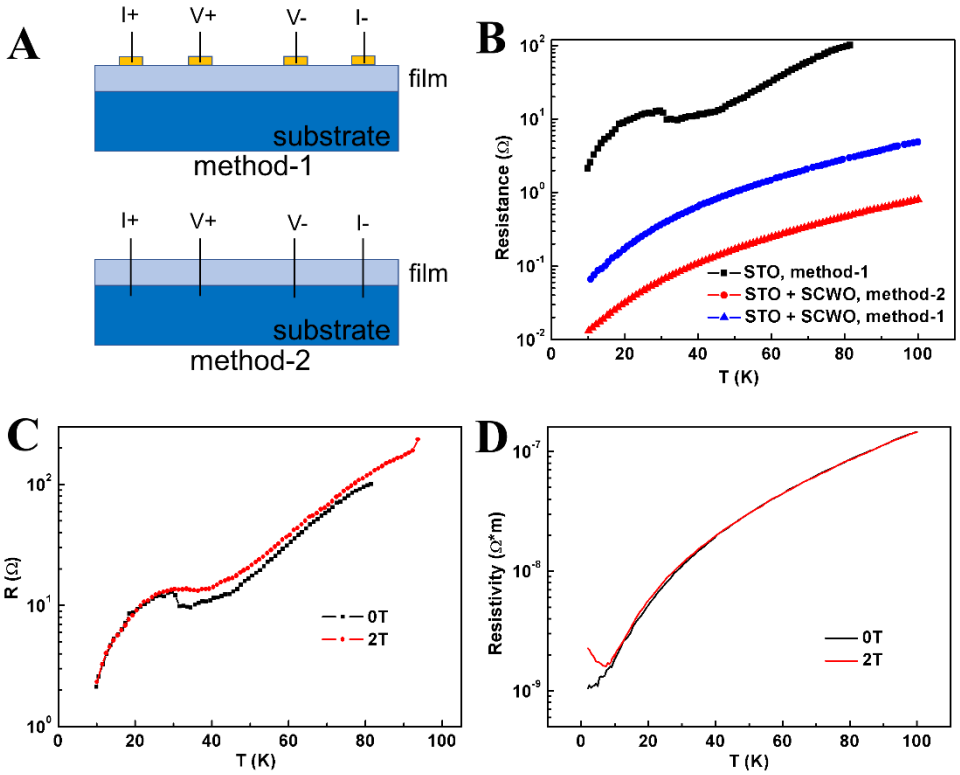

fig. S3. The methods for transport property measurement and the transport properties of the heated bare STO substrate and S-2 films with and without a magnetic field. (**A**) The two methods used to measure the transport and magnetoresistance. In both the method-1 and method-2, the magnetic field is out of plane and perpendicular to the thin film and thus perpendicular to the in-plane current. The geometry size for each measurement is constant. (**B**) Temperature-dependent resistances of heated STO substrate measured with method-1 and the S-2 films measured with both method-1 and method-2. No magnetic field is applied during these measurements. (**C**) Temperature-dependent resistances of heated STO substrate with zero field and 2 T magnetic field. Almost negligible magnetoresistance can be observed. (**D**) Temperature-dependent resistances of the S-2 films with zero field and 2 T magnetic field. Giant positive magnetoresistance is observed at low temperature range.

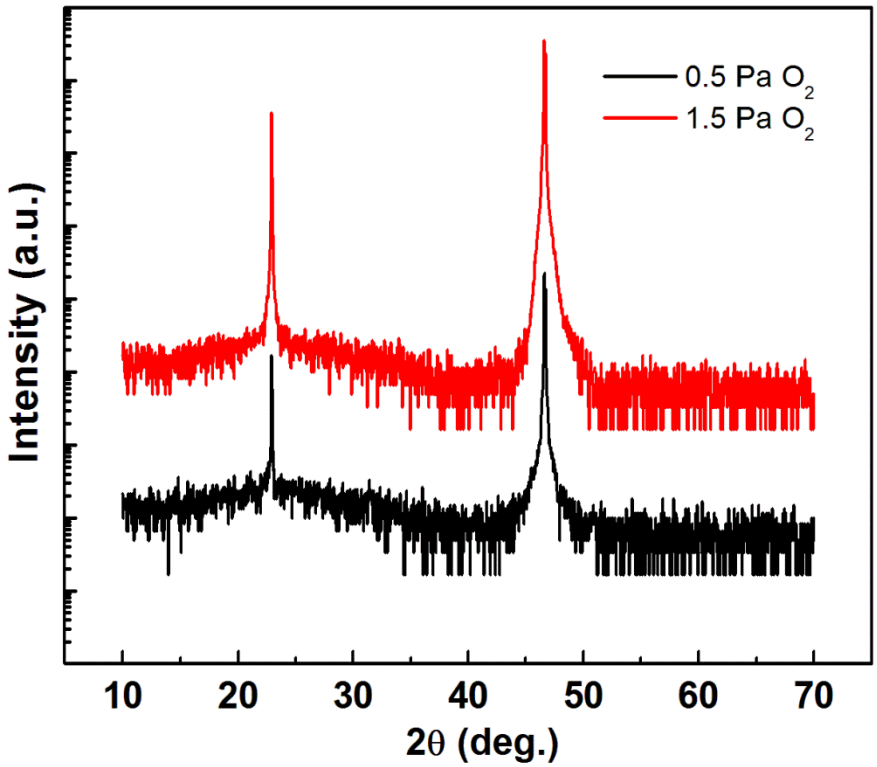

fig. S4. XRD patterns of the SCWO films fabricated with oxygen pressures of 0.5 and 1.5 Pa. Clearly, the XRD indicates the films are well *c*-axis oriented.

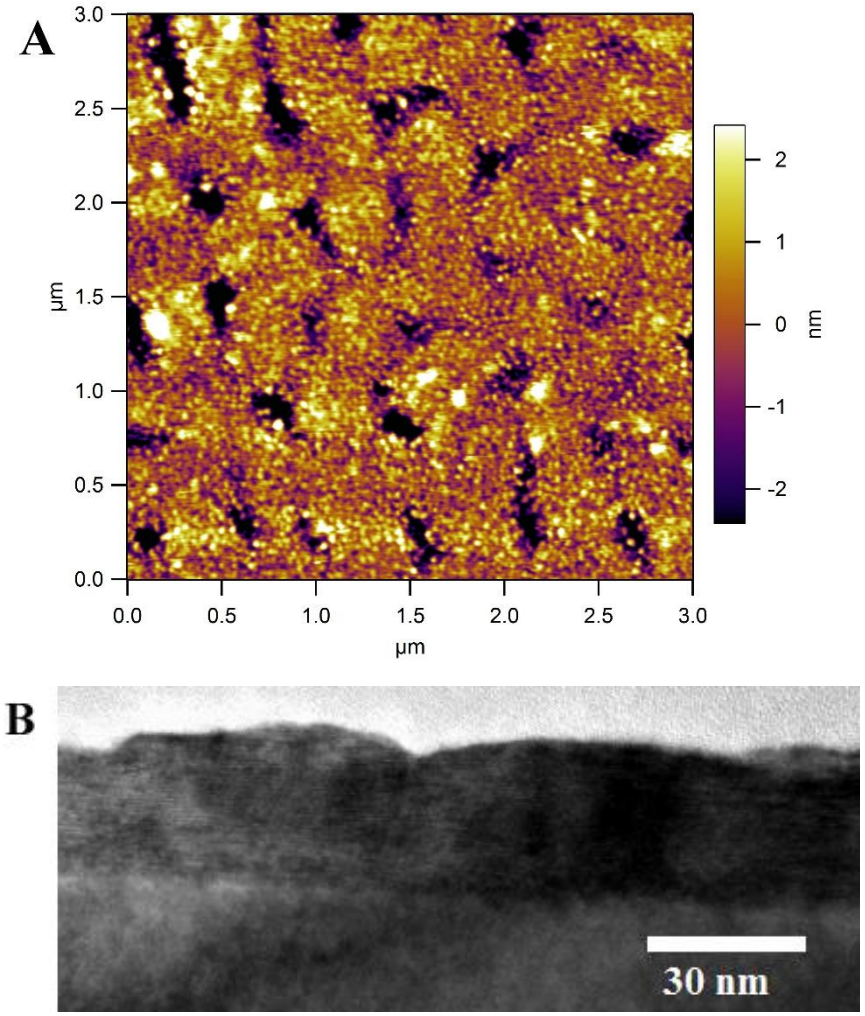

fig. S5. AFM and cross-sectional TEM images of the SCWO films fabricated with an oxygen pressure of 0.5 Pa. The films show rough surface morphology.

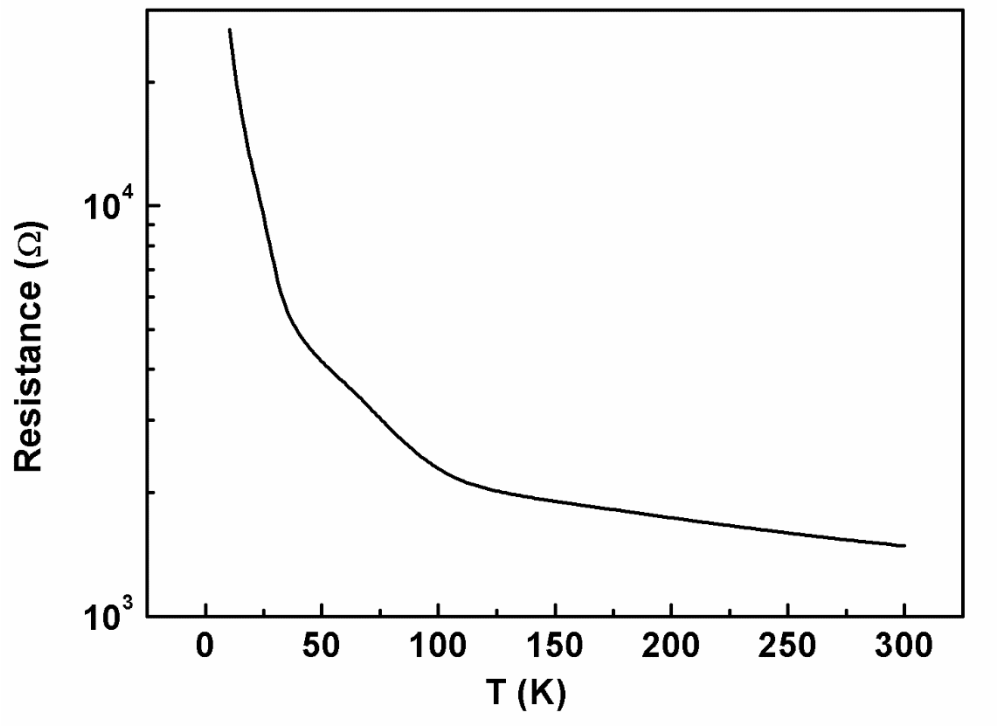

fig. S6. The temperature-dependent resistance of the films deposited under an oxygen pressure of 0.5 Pa. It shows semiconductor-like behavior in the whole temperature range.

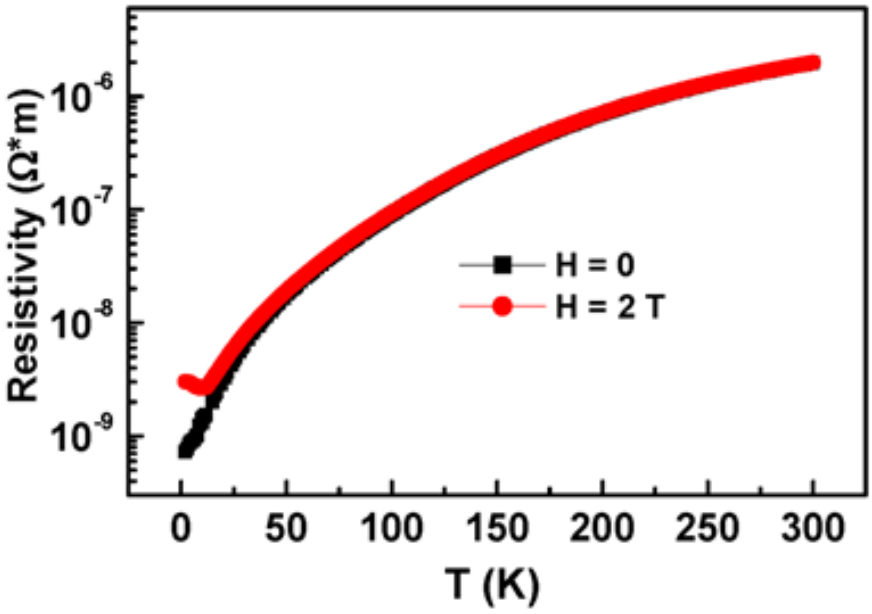

fig. S7. The temperature-dependent resistivity of the S-2 films with the external magnetic field parallel to the thin film but still perpendicular to the current. The low temperature positive MR is about 235%, very close to the MR shown in Fig. 3B, indicating the positive MR has no obvious dependence on magnetic field direction.

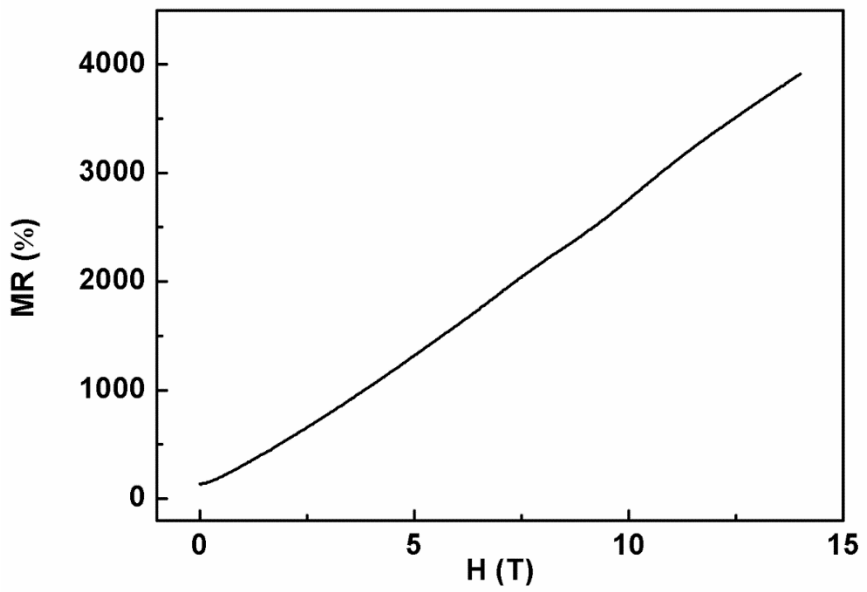

fig. S8. Field-dependent MR of the S-2 films up to 14 T. The MR shows no saturation trace.

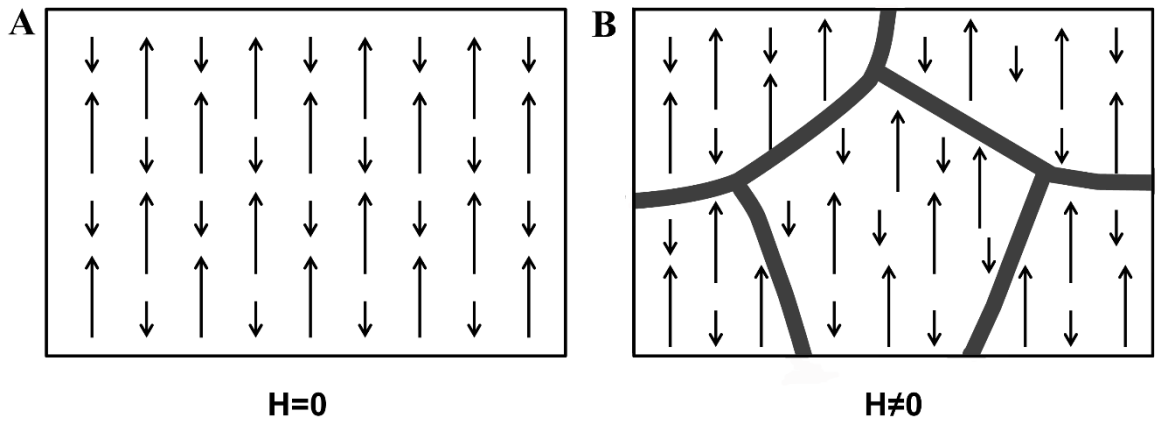

fig. S9. Evolution of magnetic structure and magnetic domain of the SCWO without and with a magnetic field. (**A**) the ideal long-range antiferromagnetic coupling between Cr^{3+} and W^{5+}, leading to macroscopic ferrimagnetic behavior with net magnetization of 2 μ_B/unit. (**B**) When an external magnetic field is applied, the long-range antiferromagnetic coupling is suppressed to form short-range spin fluctuations, such spin fluctuated regions can enhance electronic scattering, give rise to positive MR.

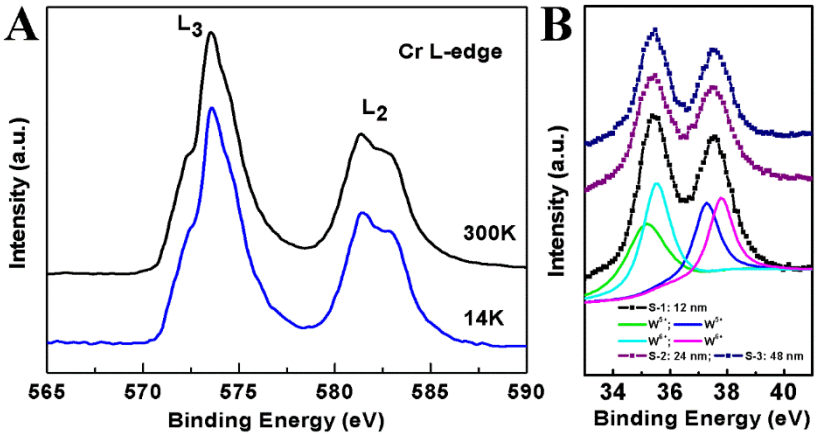

fig. S10. The valence states of Cr and W cations. (**A**) XAS of the S-2 films, confirming the coexistence of Cr^{2+} and Cr^{3+}. (**B**) W 4*f* XPS of the SCWO films, the asymmetric peaks and simulations confirm the coexistence of W^{5+} and W^{6+}.