

## Electronic Supplementary Information (ESI)

### **Rocksalt-type heavy rare earth monoxides TbO, DyO, and ErO exhibiting the metallic electronic states and ferromagnetism**

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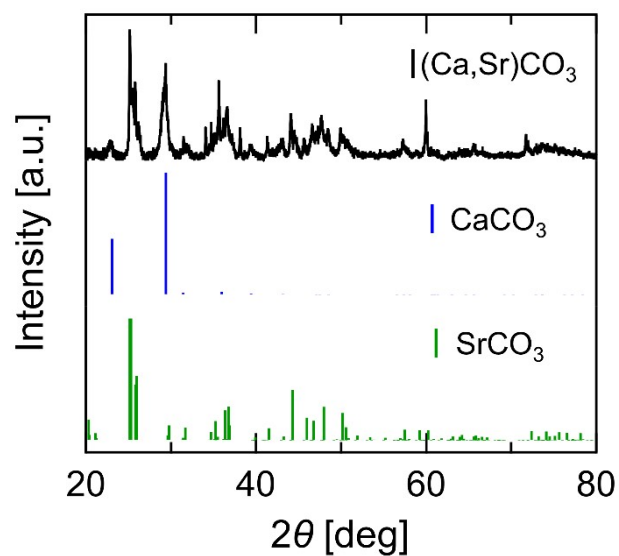
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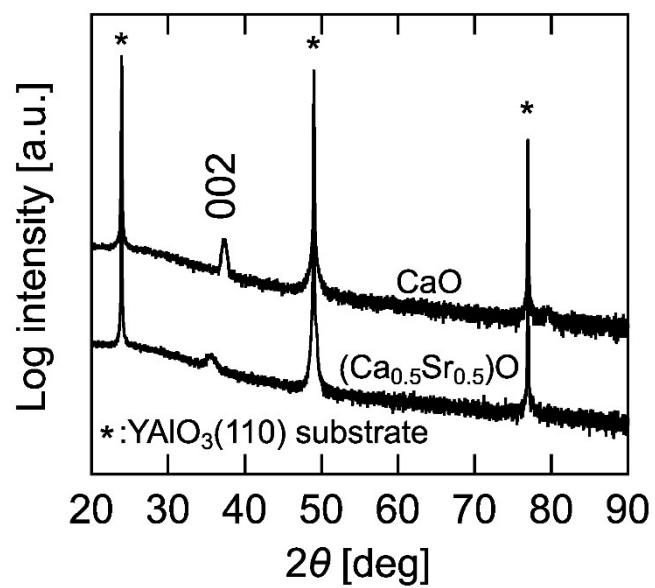
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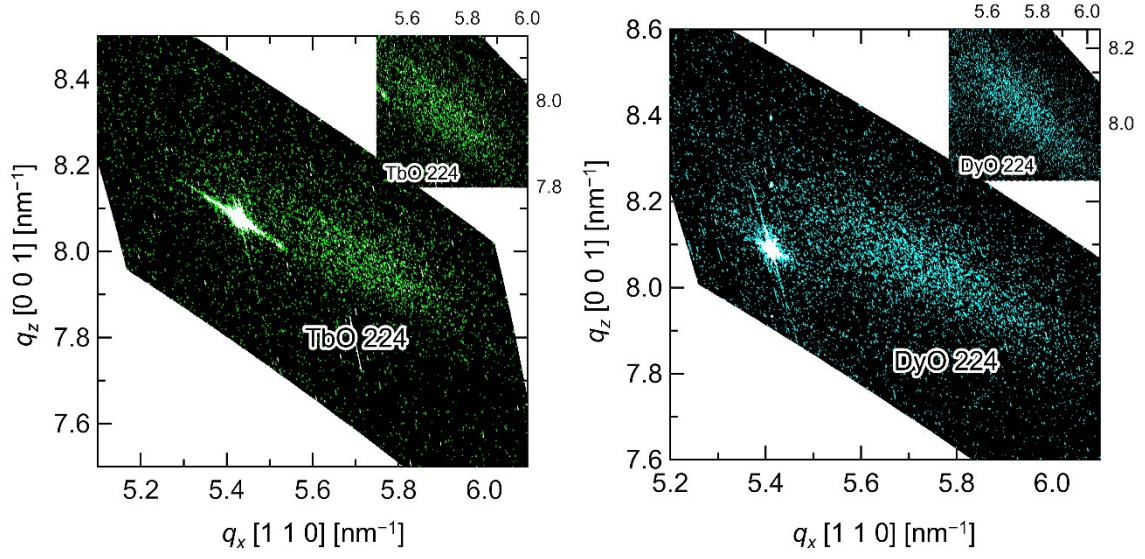
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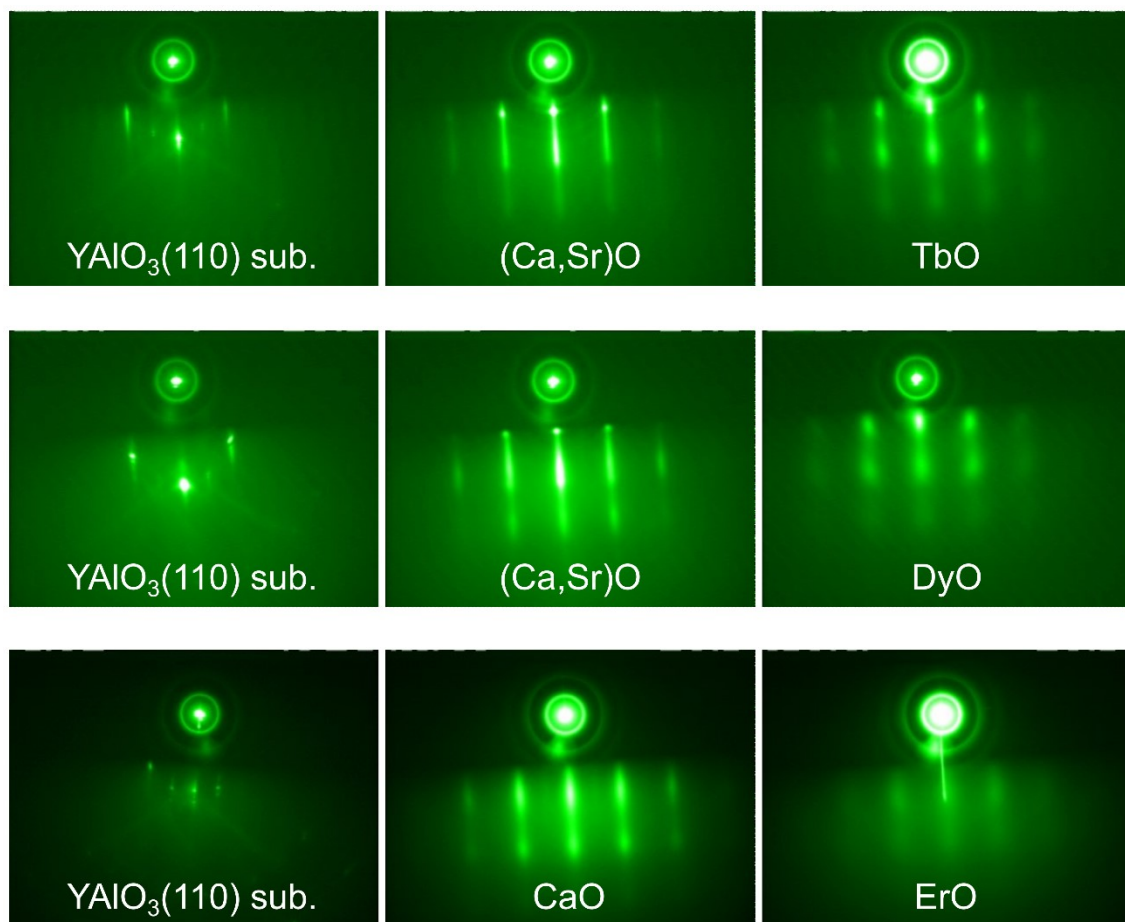
**Fig. S1** XRD  $\theta$ - $2\theta$  pattern of spark plasma sintered (Ca<sub>0.5</sub>Sr<sub>0.5</sub>)CO<sub>3</sub> target with the powder XRD pattern of CaCO<sub>3</sub> (space group: R-3cH, ICSD No. 18164) and SrCO<sub>3</sub> (space group: Pmcn, ICSD No.15195) calculated by VESTA.<sup>1</sup> Mixture of CaCO<sub>3</sub> and SrCO<sub>3</sub> powder with the molar ratio of 1:1 was pressed, and then sintered at 1200 °C under 50 MPa for 30 minutes.



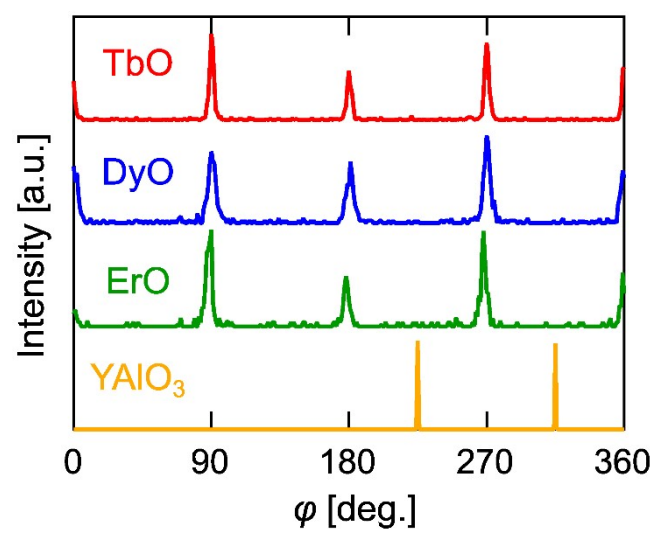
**Fig. S2** XRD  $\theta$ - $2\theta$  patterns of  $(\text{Ca}_{0.5}\text{Sr}_{0.5})\text{O}$  and  $\text{CaO}$  thin films with the same thickness as those of buffer layers for the *REO* thin films in Table 1.



**Fig. S3** Reciprocal space maps for TbO (left) and DyO (right) thin films around the *REO* 224 and  $\text{YAlO}_3$  334 diffraction peaks. Insets show the magnified images around *REO* 224 peaks.



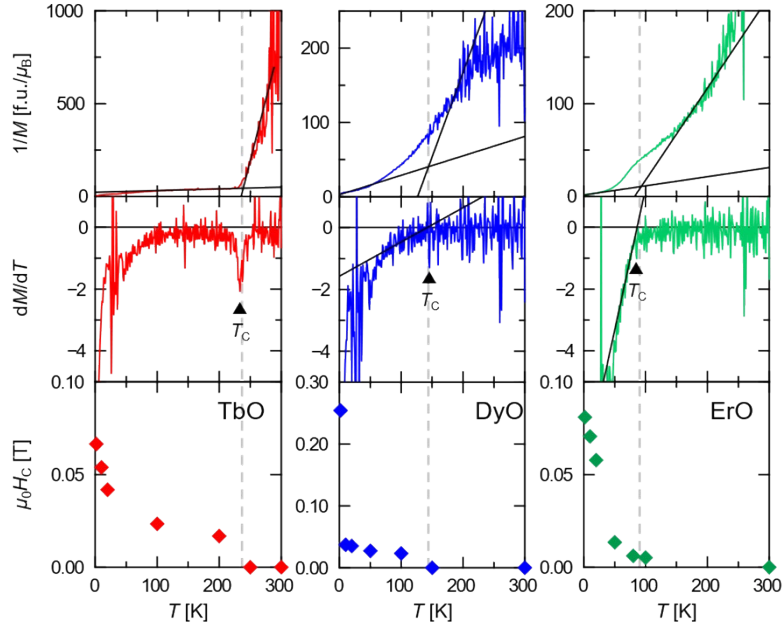
**Fig. S4** *In-situ* RHEED patterns of YAlO<sub>3</sub> substrates, (Ca,Sr)O and CaO buffer layers, and TbO, DyO, and ErO thin films along [010] during the deposition (see Fig. 1).



**Fig. S5**  $\phi$  scans taken for TbO (111), DyO (111), ErO (111), and YAlO<sub>3</sub> (101) peaks.

**Table S1** The  $3d_{5/2}$  binding energy (in the unit of eV) of Tb and Dy metals, monoxides and sesquioxides.<sup>2-4</sup>

<i>RE</i> element	<i>RE</i> metal	<i>REO</i>	<i>RE</i> <sub>2</sub> O <sub>3</sub>
Tb	1239.4	1240.4	1241.2
Dy	1293.3	1295.3	1296.5



**Fig. S6** Temperature dependences of (top panels)  $M^{-1}$ , (middle panels)  $dM/dT$  under field-cooling at 0.1 T along in-plane from red curves in Fig. 4a–c, and (bottom panels) coercive force ( $H_c$ ) evaluated from magnetic hysteresis loops in Fig. 4d–f for the REO thin films. The  $T_C$  was evaluated as the local minimum of  $dM/dT$  for TbO and the onset of  $dM/dT$  for DyO and ErO, being consistent with crossing points of linear extrapolation curves (black curves) in  $M^{-1}$ – $T$  curves.



## References

1. K. Momma, and F. Izumi, *J. Appl. Cryst.* 2011, **44**, 1272–1276.
2. B. D. Padalia, W. C. Lang, P. R. Norris, L. M. Watson and D. J. Fabian, *Proc. R. Soc. London. A. Math. Phys. Sci.*, 1977, **354**, 269–290.
3. D. J. Morgan, *Surf. Sci. Spectra*, 2023, **30**, 024017.
4. D. Barreca, A. Gasparotto, A. Milanov, E. Tondello, A. Devi and R. A. Fischer, *Surf. Sci. Spectra*, 2007, **14**, 52–59.