Supporting Information

Enhanced Curie temperature near 300 K in highly crystalline GdO epitaxial thin films concomitant with anomalous Hall effect

Takato Fukasawa,^a Dai Kutsuzawa,^b Daichi Oka,^{*ac} Kenichi Kaminaga,^d Daichi Saito,^a

Hirokazu Shimizu,^a Hiroshi Naganuma^{ef} and Tomoteru Fukumura^{*afg}

- Department of Chemistry, Graduate School of Science, Tohoku University, Sendai
 980-8578, Japan. E-mail: tomoteru.fukumura.e4@tohoku.ac.jp
- Department of Chemistry, Graduate School of Science, The University of Tokyo, Tokyo 113-0033, Japan
- c. Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University, Tokyo 192-0397, Japan. E-mail: daichi.oka@tmu.ac.jp
- d. Department of Applied Chemistry, School of Engineering, Tohoku University, Sendai 980-8579, Japan
- e. Center for Innovative Integrated Electronic Systems, Tohoku University, Sendai 980-8572, Japan
- f. Center for Science and Innovation in Spintronics, Organization for Advanced Studies, Tohoku University, Sendai 980-8577, Japan
- g. WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan



Fig. S1. Temperature derivative of magnetization as a function of temperature under the field cooling at 0.1 T for GdO (111)/CaO (111) epitaxial thin film on CaF_2 (111) substrate. The red dashed line is a guide for the eye.



Fig. S2. X-ray magnetic circular dichroism spectra for the GdO (111)/CaO (111) epitaxial thin film on CaF_2 substrate around the Gd $M_{4,5}$ edges at 70 K.



Fig. S3. (a) Temperature dependence of magnetization under the field cooling at 0.1 T for GdO (111)/CaO (111) and CaO (111) epitaxial thin films on YSZ (111) substrates. The sudden change around 100 K in the former is the noise derived from the machine error. (b) Kouvel-Fischer plot of magnetization M as a function of temperature T for GdO (111)/CaO (111) epitaxial thin film on YSZ (111) substrate. The linear fitting was performed for the data points from 270 K to 290 K (dashed red line).



Fig. S4. (a)–(c) Direction of magnetic moments (red and blue vectors) in GdO (111) thin film under different magnetic fields, (d)–(f) their projection on the (111) plane, and (g)–(h) corresponding magnetic domain structure under (a),(d),(g) zero, (b),(e),(h) low, and (c),(f),(i) high magnetic field, where the magnetization easy axes are supposed to be along the <100> directions. (g) Under the zero field, a conventional 180° domain with straight domain walls is formed. (h) Under the low field, the domain walls become zigzag shape due to the magnetization rotation from the $[0\bar{1}0]$ direction (shown in (a)) to another easy axis of either the [100] or [010] direction (shown in (b)), resulting in positive magnetoresistance caused by of the carrier scattering at the increased domain walls. (i) Under the high field, single magnetic domain is formed, resulting in negative magnetoresistance.