ADVANCED MATERIALS

Supporting Information

for *Adv. Mater.,* DOI: 10.1002/adma.202200626

Realization of a Half Metal with a Record-High Curie Temperature in Perovskite Oxides

Zhehong Liu, Shuaikang Zhang, Xiao Wang, Xubin Ye, Shijun Qin, Xudong Shen, Dabiao Lu, Jianhong Dai, Yingying Cao, Kai Chen, Florin Radu, Wen-Bin Wu, Chien-Te Chen, Sonia Francoual, José R. L. Mardegan, Olaf Leupold, Liu Hao Tjeng, Zhiwei Hu, Yi-feng Yang, and Youwen Long**

Supporting Information

Realization of a half metal with a record-high Curie temperature in perovskite oxides

Zhehong Liu, Shuaikang Zhang, Xiao Wang, Xubin Ye, Shijun Qin, Xudong Shen, Dabiao Lu, Jianhong Dai, Yingying Cao, Kai Chen, Florin Radu, Wen-Bin Wu, Chien-Te Chen, Sonia Francoual, José R. L. Mardegan, Olaf Leupold, Liu Hao Tjeng, Zhiwei Hu, Yi-feng Yang, Youwen Long**

Dr. Z. Liu, Dr. S. Zhang, Dr. X. Ye, Dr. S. Qin, Dr. X. Shen, Dr. D. Lu, Dr. J. Dai, Dr. Y. Cao, Prof. Y. Yang, Prof. Y. Long

Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

E-mails: ywlong@iphy.ac.cn; yifeng@iphy.ac.cn

Dr. Z. Liu, Dr. S. Zhang, Dr. X. Ye, Dr. S. Qin, Dr. D. Lu, Dr. J. Dai, Dr. Y. Cao, Prof. Y. Yang, Prof. Y. Long

School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China

Dr. X. Wang, Prof. L. Tjeng, Dr. Z. Hu,

Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany

Dr. X. Shen, Prof. Y. Yang, Prof. Y. Long

Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China

Dr. K. Chen, Dr. F. Radu

Helmholtz-Zentrum Berlin fur Materialien und Energie, Albert-Einstein-Str.15, 12489 Berlin, Germany

Dr. W. Wu, Prof. C. Chen

National Synchrotron Radiation Research Center, Hsinchu 30076, Taiwan

Dr. S. Francoual, Dr. O. Leupold, Dr. J. Mardegan

Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany

*Corresponding author.

Supporting Information 1. Density of states and band structure results of $LaCu_3Fe_2Re_2O_{12}$

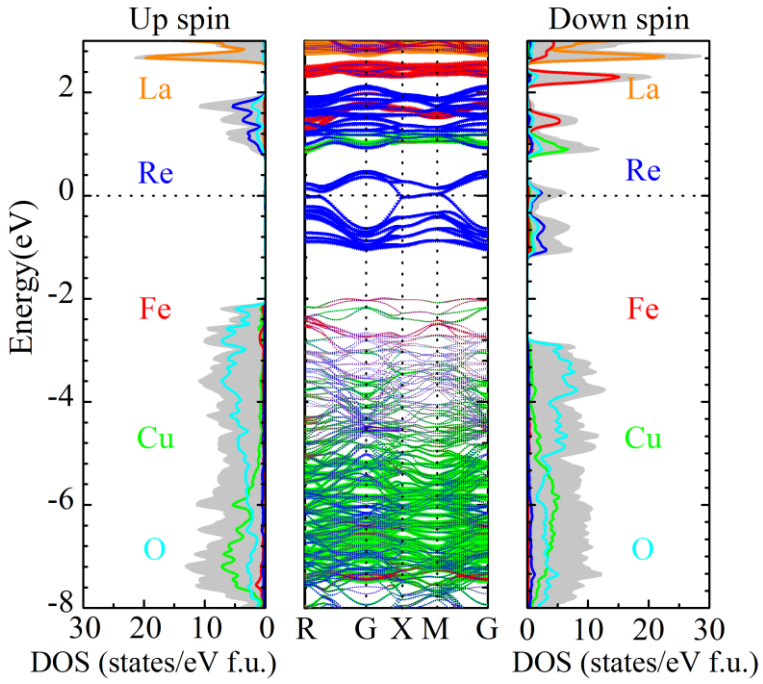

Figure S1. Density of states and band structure results of $LaCu_3Fe_2Re_2O_{12}$ using GGA+U+SOC. The up-spin energy gap increases from 2.27 eV in GGA+U to 2.62 eV in the current GGA+U+SOC.

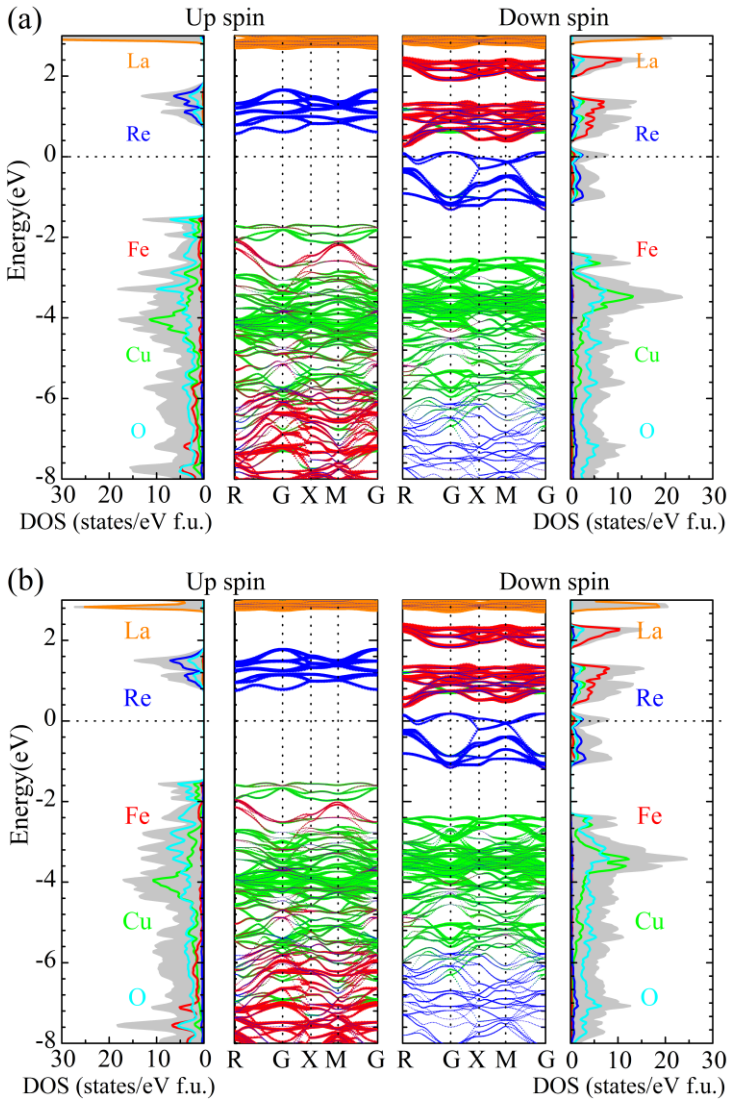

Figure S2. Density of states and band structure results of $LaCu_3Fe_2Re_2O_{12}$ when (a) compression, and (b) tensile strain by 2% were applied. The half-metallic bandgap slightly decreases to 2.26 eV and increases to 2.30 eV in these two cases from the ambient 2.27 eV, respectively.

Supporting Information 2. Refined structure parameters and BVS results

Table S1. Refined structure parameters of LCFRO and the BVS results for Cu and Fe at room temperature. The BVS values (V_i) of different ions were calculated according to the formula $V_i = \sum_j S_{ij}$, and $S_{ij} = \exp[(r_0-r_{ij})/0.37]$. The value of r_0 is 1.679 for Cu, 1.759 for Fe. For the *A*'-site Cu, twelve coordinated oxygen atoms were used, and for the *B*-site Fe, six coordinated oxygen atoms were used in the BVS calculation.

Parameter	LCFRO
a (Å)	7.4988(1)
O_x	0.5726(5)
O_y	0.7550(3)
O_z	0.0555(5)
U_{iso} (La) (100×Å^2)	2.67(5)
U_{iso} (Cu) (100× Å^2)	2.53(5)
U_{iso} (Fe) (100× Å^2)	1.62(4)
U_{iso} (Re) (100× Å^2)	1.85(1)
U_{iso} (O) (100× Å^2)	0.7(1)
Cu-O (×4)	1.975(3)
Fe-O (×6)	2.032(2)
Re-O (×6)	1.960(2)
∠Fe-O-Re (deg)	139.9(2)
∠Cu-O-Re (deg)	111.2(1)
∠Cu-O-Fe (deg)	108.3(2)
BVS (Cu)	2.02
BVS (Fe)	2.88
R_{wp} (%)	3.93
R_p (%)	2.56

Supporting Information 3. Orbital and spin moments of transition-metal ions

Table S2. Orbital and spin moments of transition-metal ions of LCFRO. Considering N_d = 9, 5, 2.5 for Cu^{2+}, Fe^{3+} and $Re^{4.5+}$ ions, respectively, orbital and spin moments of Cu^{2+}, Fe^{3+} and $Re^{4.5+}$ in LCFRO can be calculated by sum rules[40,41]. Here, N_d denotes the electron occupation number. The XMCD of Cu^{2+} and Fe^{3+} ($Re^{4.5+}$) ions were measured at 6 T, 50 K (5 T, 5 K). Since the Re 5d t_{2g} subshell is less than half-filled, the sign for spin and orbital moment is opposite, following the Hund's Rules.

Ions	M_{orb} (μ_B/atom)	M_{spin} (μ_B/atom)	M_{orb}/M_{spin}
Cu^{2+}	0.05	0.57	0.09
Fe^{3+}	0.04	3.94	0.01
$Re^{4.5+}$	0.36	−1.36	-0.26

Supporting Information 4. Calculated formation energies of $LaCu_3Fe_2Re_2O_{12}$ and its' starting materials

Table S3. Formation energies of $LaCu_3Fe_2Re_2O_{12}$ and its' starting materials.

	E (eV)
La_2O_3	-468631.4888
CuO	-47080.88048
Fe_2O_3	-75406.30069
Re_2O_7	-924195.033
Re	-454928.0502
$LaCu_3Fe_2Re_2O_{12}$	-1370058.169
ΔE	-19.49735873