Supporting Information

For

High-pressure synthesis and high-performance half metallicity of quadruple perovskite oxide $DyCu_3Fe_2Re_2O_{12}$

Zhehong Liu[a,b,†], Jinfeng Peng[a,†], Xiao Wang[a], Fedor Temnikov[c], Alexey Ushakov[c], Xubin Ye[a,b], Zhao Pan[a,b], Jie Zhang[a,b], Maocai Pi[a,b], Shuai Tang[a,b], Kai Chen[d], Florin Radu[e], Zhiwei Hu[f], Chien-Te Chen[g], Zhenhua Chi[h], Zlata Pchelkina[c], Valentin Irkhin[c], Sergey V. Streltsov[c,i,] *, Youwen Long[a,b,j,]*

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

[b]School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China

[c]Institute of Metal Physics, S. Kovalevskaya Street 18, 620108 Ekaterinburg, Russia

[d]National Synchrotron Radiation Laboratory, University of Science and Technology of China, Hefei 230026, China

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

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

[g]National Synchrotron Radiation Research Center (NSRRC), Hsinchu Science Park, Hsinchu 300092, Taiwan

[h]Institute of Plasma Physics, HFIPS, Chinese Academy of Sciences, Hefei, 230031, China

[i]Department of Theoretical Physics and Applied Mathematics, Ural Federal University, Mira St. 19, 620002 Ekaterinburg, Russia

[j]Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China

*Corresponding emails: streltsov.s@gmail.com; ywlong@iphy.ac.cn

[†]Equally contributed to this work.

Tab. S1. Refined structure parameters of CCCRO and the calculated BVS results of 3*d* transition metal Cu and Fe at RT. 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	DCFRO
a (Å)	7.44611(1)
O_x	0.5726(5)
O_y	0.7550(3)
O_z	0.0555(5)
U_{iso}(Dy) (100×Å^2)	3.56(7)
U_{iso}(Cu) (100×Å^2)	1.04(6)
U_{iso}(Fe) (100×Å^2)	0.32(5)
U_{iso}(Re) (100×Å^2)	0.80(3)
U_{iso}(O) (100×Å^2)	0.4(1)
Cu-O (×4)	1.961(5)
Cu-O (×4)	2.805(7)
Cu-O (×4)	3.308(9)
Fe-O (×6)	2.017(5)
Re-O (×6)	1.947(4)
∠Fe-O-Re (deg)	139.9(1)
∠Cu-O-Re (deg)	111.2(1)
∠Cu-O-Fe (deg)	108.3(1)
BVS (Cu)	2.11
BVS (Fe)	2.99
R_{wp} (%)	4.47
R_p (%)	2.66

Tab. S2. Orbital and spin moments of transition-metal ions and rare-earth ion of DCFRO. Considering N_d = 9, 5, 5 for Cu^{2+}, Fe^{3+} and Dy^{3+} ions, respectively, orbital and spin moments of Cu^{2+}, Fe^{3+} and Dy^{3+} in CCFRO can be obtained by the XMCD sum rules [27,28]. Here, N_d denotes the electron occupation number. XMCD of Cu^{2+}, Fe^{3+} and Dy^{3+} ions were measured at 6 T, 8 K.

Ions	M_{orb} (μ_B/atom)	M_{spin} (μ_B/atom)	M_{orb}/M_{spin}
Cu^{2+}	0.14	0.49	0.29
Fe^{3+}	0.04	4.12	0.02
Dy^{3+}	2.12	1.60	1.33

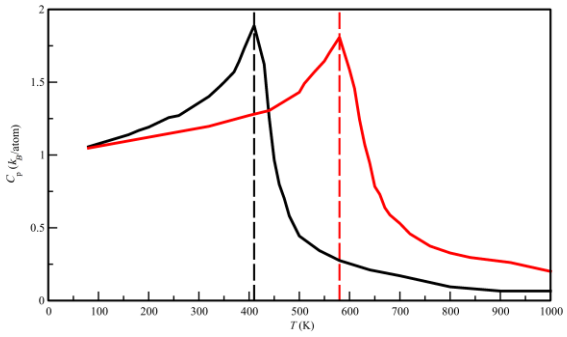

Fig. S1. Specific heat of $DyCu_3Fe_2Re_2O_{12}$ calculated using the classical Monte Carlo method. The black curve is based on theoretically calculated exchange parameters. The red curve also takes into account quantum renormalization factor ($S^2 \rightarrow S(S+1)$).