Supplemental Material: Ferroic order for anisotropic magnetic dipole term in collinear antiferromagnets of $(t_{2g})^4$ system

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S1. ANISOTROPIC MAGNETIC DIPOLE OPERATOR

The anisotropic magnetic dipole (AMD) moment $\boldsymbol{t}$ of a particle electron state with orbital l and spin s , i.e., $|\phi\rangle = \sum_{\langle l_z,s_z\rangle} a_{l_z,s_z}|l,l_z,s,s_z\rangle$, is defined as the expectation value of the intra-atomic magnetic dipole operator $\langle \boldsymbol{t}\rangle = \langle\phi|\hat{\boldsymbol{t}}|\phi\rangle$ [1]; l_z and s_z represent the orbital and spin quantum numbers, respectively, and a_{l_z,s_z} is a normalization factor. The AMD operator t_α ($\alpha = x, y, z$) is expressed as

$$t_\alpha = [\boldsymbol{s} - 3(\hat{r}\cdot\boldsymbol{s})\hat{r}]_\alpha = \sum_{\beta=x,y,z} Q_{\alpha\beta}s_\beta, \tag{1}$$

where $\boldsymbol{s}$ and $\hat{r}$ are the operators for spin and the unit vector of position, respectively, and $Q_{\alpha\beta}$ denotes the electric quadrupole operators [2, 3]. The t_z term for d_{xy}, d_{xy}, d_{zx}, $d_{x^2-y^2}$, and $d_{3z^2-r^2}$ are 2/7, -1/7, -1/7 , 2/7, and -2/7, respectively.

Next, $\mathcal{PT}$ symmetry for the AMD moment is discussed. The magnetic space group of antiferromagnetism ordered is $Pnn'm'$ when $\boldsymbol{N}$||[100] of the rutile structure, including the magnetic symmetric operations $\mathcal{P}$, $\mathcal{T}\mathcal{M}_x\boldsymbol{\tau}_{1/2}$, $\mathcal{M}_y\boldsymbol{\tau}_{1/2}$, and $\mathcal{T}\mathcal{M}_z$ [4]. Here, $\mathcal{T}$, $\mathcal{P}$, $\mathcal{M}_\alpha$ (α=x, y, z), and $\boldsymbol{\tau}_{1/2}$ denote the time reversal, spatial inversion, mirror perpendicular to the α-axis, and translation of the $(1/2, 1/2, 1/2)$ operations, respectively. Those operations for the AMD moment $\boldsymbol{t}$ result in $\mathcal{T}\boldsymbol{t} = -\boldsymbol{t}$, $\mathcal{P}\boldsymbol{t} = \boldsymbol{t}$, and $\boldsymbol{\tau}_{1/2}\boldsymbol{t} = \boldsymbol{t}$. The mirror operation is expressed as

$$\mathcal{M}_y\boldsymbol{t} = (-t_x, t_y, -t_z), \tag{2}$$

indicating that all components other than the component perpendicular to the mirror are inverted. Thus, it can be confirmed that $\boldsymbol{t}$||[010] is allowed from the viewpoint of magnetic symmetry. When the vector $\boldsymbol{N}$ is inverted, $\langle\boldsymbol{t}\rangle$ is inverted.

S2. Model Hamiltonian

For the Ru^{4+} model describing an electronic state of RuO_2, Hamiltonian is given by

$$H_i = H_{\rm atom} + H_{\rm CEF} + H_{\rm MF}, \tag{3}$$

where index i indicates Ru1 and Ru2 shown in Fig. 1(a) of the main text. The first term $H_{\rm atom}$ is expressed as

$$\begin{aligned} H_{\rm atom} = \;& \epsilon_d \sum_\gamma d^\dagger_\gamma d_\gamma + \zeta_{4d}\sum_{\gamma_1,\gamma_2}(\boldsymbol{l}\cdot\boldsymbol{s})_{\gamma_1,\gamma_2} d^\dagger_{\gamma_1} d_{\gamma_2} \\ & + \epsilon_p \sum_\gamma p^\dagger_\gamma p_\gamma + \zeta_{2p}\sum_{\gamma_1,\gamma_2}(\boldsymbol{l}\cdot\boldsymbol{s})_{\gamma_1,\gamma_2} p^\dagger_{\gamma_1} p_{\gamma_2} \\ & + \frac{1}{2}\sum_{\gamma_1,\gamma_2,\gamma_3,\gamma_4} g_{dd}(\gamma_1,\gamma_2;\gamma_3,\gamma_4)\, d^\dagger_{\gamma_1} d_{\gamma_2} d^\dagger_{\gamma_4} d_{\gamma_3} \\ & + \sum_{\gamma_1,\gamma_2,\gamma_3,\gamma_4} g_{dp}(\gamma_1,\gamma_2;\gamma_3,\gamma_4)\, d^\dagger_{\gamma_1} p^\dagger_{\gamma_2} p_{\gamma_4} d_{\gamma_3}, \end{aligned} \tag{4}$$

where $d^\dagger_\gamma$ represents the creation operator for a $4d$ electron, including a combined index γ with orbital and spin, and $p^\dagger_\gamma$ represents the creation operator for a $2p$ core state [5–8]. $H_{\rm atom}$ includes $4d$ level (ϵ_d), spin-orbit coupling constant for the $4d$ orbital (ζ_{4d}), $2p$ level (ϵ_p), spin-orbit coupling constant for the $2p$ orbital (ζ_{2p}), Coulomb interaction between the $4d$ states (g_{dd}), and the Coulomb interaction between the $4d$ and $2p$ states (g_{dp}). These spin-orbit coupling constants and the Slater integrals included in g_{dd} and g_{dp} are estimated from the ionic calculation within the Hartree–Fock–Slater (HFS) method [9]. The ζ_{4d} of Ru^{4+} is 0.161 eV, and more we investigated spin-orbit interaction (SOI) dependence of the electronic state by changing ζ_{4d}. For the Slater integrals, 60% of the HFS values are used [10, 11]. The second term $H_{\rm CEF}$ is determined by considering the one-electron potential of D_{2h} symmetry [11], expressed as

$$\begin{aligned} V_{\rm crys} = \;& B^2_0 C^{(2)}_0 + B^2_2(C^{(2)}_2 + C^{(2)}_{-2}) \\ & + B^4_0 C^{(4)}_0 + B^4_2(C^{(4)}_2 + C^{(4)}_{-2}) + B^4_4(C^{(4)}_4 + C^{(4)}_{-4}), \end{aligned} \tag{5}$$

where

$$C^{(k)}_q = \sqrt{\frac{4\pi}{2k+1}}\, Y_{kq}, \tag{6}$$

and Y_{kq} represents a spherical harmonic [12]. Considering the previous study in Ref. 11, the $10Dq$ value between e_g ($d_{3z^2-r^2}/d_{xy}$) and t_{2g} ($d_{x^2-y^2}/d_{yz}/d_{zx}$) is 2.6 eV and the e_g splitting between d_{xy} and $d_{3z^2-r^2}$ is 0.6 eV. For the splitting of t_{2g} orbitals in the present study, Δ for the splitting between $d_{x^2-y^2}$ and d_{yz}/d_{zx} is 1.0 eV and the splitting between d_{yz} and d_{zx} is 0.55 eV. The third term $H_{\rm MF}$ is expressed as

$$H_{\rm MF} = \sum_{\gamma_1,\gamma_2}(\boldsymbol{h}^{(i)}_{\rm MF}\cdot\boldsymbol{s})_{\gamma_1,\gamma_2}\, d^\dagger_{\gamma_1} d_{\gamma_2}, \tag{7}$$

where $\boldsymbol{h}^{(i)}_{\rm MF}$ denotes the molecular field for the spin part of the Ru1 and Ru2.

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