Supporting Information

A Highly Anisotropic Cobalt (II) Based Single Chain Magnet: Exploration of Spin-Canting in an Antiferromagnetic Array

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Transformation from the spin operators $s_\gamma(A)$, $s_\gamma(B)$ defined in the molecular frame to the operators s_{γ_A} , s_{γ_B} defined in the local frames

$$\begin{pmatrix} s_X(A) \\ s_Y(A) \\ s_Z(A) \end{pmatrix} = \begin{pmatrix} \cos(\varphi/2) & 0 & \sin(\varphi/2) \\ 0 & 1 & 0 \\ -\sin(\varphi/2) & 0 & \cos(\varphi/2) \end{pmatrix} \begin{pmatrix} s_{X_A} \\ s_{Y_A} \\ s_{Z_A} \end{pmatrix},$$

$$\begin{pmatrix} s_X(B) \\ s_Y(B) \\ s_Z(B) \end{pmatrix} = \begin{pmatrix} \cos(\varphi/2) & 0 & -\sin(\varphi/2) \\ 0 & 1 & 0 \\ \sin(\varphi/2) & 0 & \cos(\varphi/2) \end{pmatrix} \begin{pmatrix} s_{X_B} \\ s_{Y_B} \\ s_{Z_B} \end{pmatrix}. \tag{SI.1}$$

Relations between the pseudo-spin-1/2 operators $\tau_{Z_A}(i)$, $\tau_{Z_B}(i)$ defined in the local frames and the operators $\tau_X^A(i)$, $\tau_Z^A(i)$, $\tau_X^B(i)$, $\tau_Z^B(i)$ defined in the molecular frame

$$\begin{aligned} \tau_{Z_A}(i) &= \sin(\varphi/2)\,\tau_X^A(i) + \cos(\varphi/2)\,\tau_Z^A(i), \\ \tau_{Z_B}(i) &= -\sin(\varphi/2)\,\tau_X^B(i) + \cos(\varphi/2)\,\tau_Z^B(i), \end{aligned} \tag{SI.2}$$

Relations between the magnetic field components $\tau_{Z_A}(i)$, $\tau_{Z_B}(i)$ defined in the local frames and magnetic field components H_X, H_Z defined in the molecular frame

$$\begin{aligned} H_{Z_A} &= \sin(\varphi/2)\,H_X + \cos(\varphi/2)\,H_Z , \\ H_{Z_B} &= -\sin(\varphi/2)\,H_X + \cos(\varphi/2)\,H_Z . \end{aligned} \tag{SI.3}$$

Expressions for the components of the local *g*-tensors defined in the molecular frame

$$g_{ZZ}^A = g_{ZZ}^B = g_{||}\cos^2(\varphi/2), \qquad g_{XX}^A = g_{XX}^B = g_{||}\sin^2(\varphi/2), \tag{SI.4}$$

$$g_{XZ}^A = g_{ZX}^A = -g_{XZ}^B = -g_{ZX}^B = \tfrac{1}{2}\,g_{||}\sin(\varphi).$$