Supplemental Material for Relief of spin frustration due to Dzyaloshinskii–Moriya interactions in quasi-one-dimensional $S = 1/2$ antiferromagnet by magnetic anisotropy

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MAGNETIC STRUCTURE

Several magnetic reflections were detected using Sika (Fig. S1). Below T_{N}, magnetic reflections were appeared at $(\frac{n}{2}kl)$ where n is an odd integer. We further explored magnetic reflection peaks in a wide reciprocal space using SENJU. The integrated intensity decreases with increasing temperature and then disappears above 0.54 K (Fig. S2). Four magnetic Cu^{2+} ions are located on the Wyckoff positions $4c$: $\mathbf{r}_1 = (0.636, 0.25, 0.966)$, $\mathbf{r}_2 = (0.136, 0.25, 0.534)$, $\mathbf{r}_3 = (0.364, 0.75, 0.034)$, and $\mathbf{r}_4 = (0.864, 0.75, 0.466)$ in the orthorhombic unit cell. The crystal structure of $Na_2CuSO_4Cl_2$ is described by the orthorhombic space group $Pnma$, and thus there are eight irreducible representations in the magnetic unit cell. Assuming that an antiferromagnetic commensurate structure is formed, the possible magnetic structure can be represented by only four irreducible spin arrangements $(\uparrow,\uparrow,\uparrow,\uparrow)$, $(\downarrow,\uparrow,\uparrow,\uparrow)$, $(\uparrow,\downarrow,\uparrow,\uparrow)$, and $(\downarrow,\downarrow,\uparrow,\uparrow)$, where the brackets show the relative phase factors of the spins on the sites ($\mathbf{r}_1$, $\mathbf{r}_2$, $\mathbf{r}_3$, $\mathbf{r}_4$) (see Fig. 2(b)). Therefore, in the case that the spins are pointing along one of the crystal axes, there are only 12 possible magnetic structures [1]. The calculation results for the 12 magnetic structures are shown in TABLE I. Each panel shows the observed intensities $I^{\mathrm{Mag.}}_{\mathrm{obs.}}$ plotted against the calculated values $I^{\mathrm{Mag.}}_{\mathrm{calc.}}$ for the magnetic peaks. We can see that there is a significant difference in the reliability coefficient for the models, indicating the realization of the magnetic structure shown in Fig. 2(b) of the main text.

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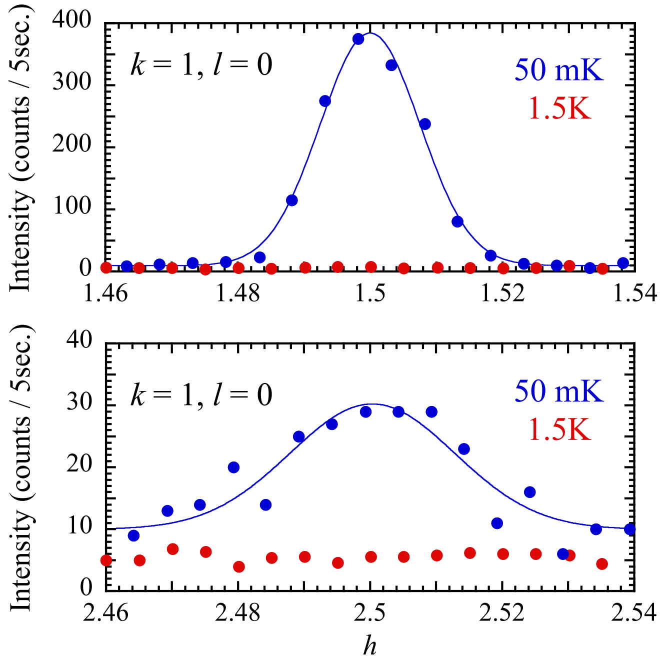

FIG. S1: Diffraction patterns taken around the $h = 1.5$ $k = 1$ $l = 0$ and $h = 2.5$ $k = 1$ $l = 0$ positions at 1.5 K and at 50 mK as measured on the Sika.

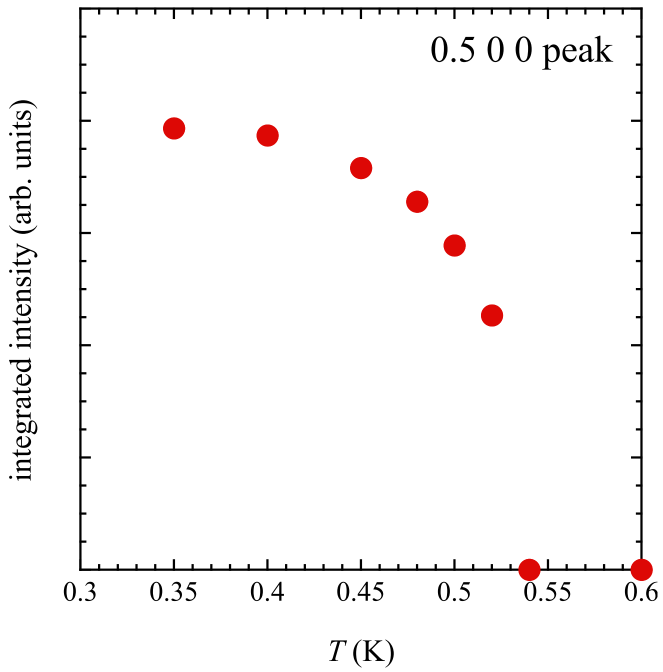

FIG. S2: Temperature dependence of the intensity of the 0.5 0 0 magnetic peak for $Na_2CuSO_4Cl_2$.

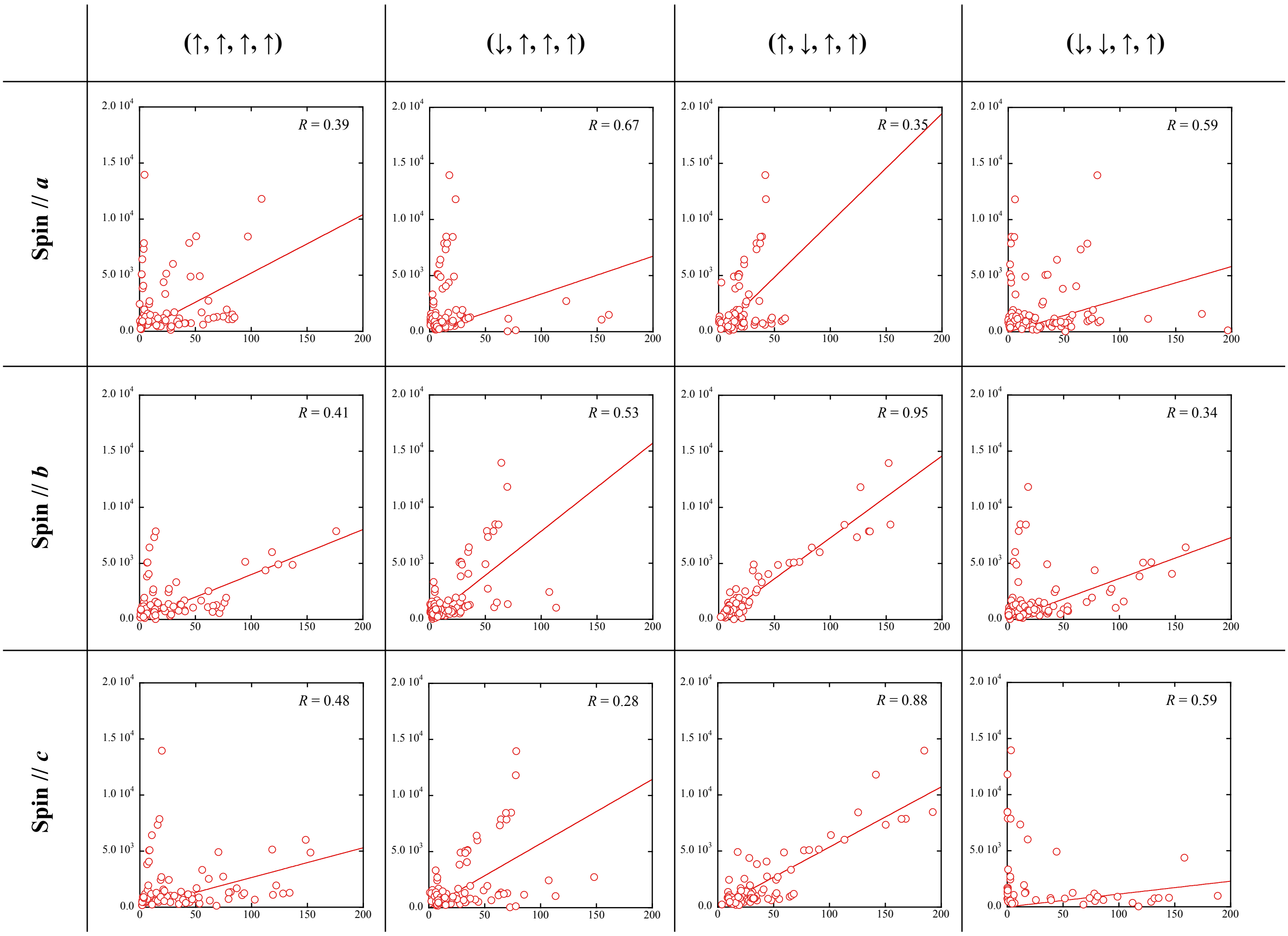

TABLE I: (Color online) Calculation results for the 12 magnetic structures of $Na_2CuSO_4Cl_2$. The values in the plot show the obtained ordered moment and Pearson's correlation coefficient R.