

## Degree of frustration in geometrically frustrated magnets $(\text{Co}_{1-x}\text{Zn}_x)\text{Cr}_2\text{O}_4$

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Highly frustrated systems, spinel antiferromagnets  $\text{ACr}_2\text{O}_4$  (nonmagnetic  $\text{A}=\text{Mg}, \text{Zn}, \text{Cd}$  and  $\text{Hg}$ ) consisting of a corner-sharing tetrahedron lattice as B (Cr) sites, are well-known to exhibit the phenomenon of spin hexamer on the kagome plane. On the other hand, it was recently reported that geometric frustration still survives in spinel ferrimagnets  $\text{ACr}_2\text{O}_4$  (magnetic  $\text{A}=\text{Co}$  and  $\text{Mn}$ ) as explained below. In these materials a spin forms a conical structure resolved into an Ising-type ferrimagnetic component and an incommensurate spiral one. As the temperature decreases, although the former component forms long-range order at  $T_C=93$  and  $51$  K, the latter one mainly starts to grow at  $T_S=25$  and  $14$  K and does not achieve long-range order even in the lowest temperature phase. Therefore, it was proposed that the geometric frustration among the B-site spins can coexist with the A-site spins, and is mainly reflected on the spiral component.

However, the spiral type seems to be too different from the hexamer. Here, in order to validate the expanded concept of geometric frustration, we report the spiral spin correlations of solid solution materials  $(\text{Co}_{1-x}\text{Zn}_x)\text{Cr}_2\text{O}_4$  by powder neutron diffraction ( $x=0$  to  $1$ ).

Figure shows the diffraction patterns measured at elastic condition in the lowest temperature phases. The intensity of 111 fundamental reflection decreases with increasing  $x$  from  $0$  to  $0.45$ , then is almost constant, meaning that the ferrimagnetic component disappears. The magnetic satellite reflections monotonically become broad, most likely validating that the correlation length of spiral order corresponds to the degree of expanded frustration, and the spiral short-range order comes from the

frustration. In addition, even at  $x=0.90$  the peak positions of diffuse scattering are different from those of magnetic Bragg reflections in  $\text{ZnCr}_2\text{O}_4$ , suggesting that the spiral short-range order still survives.

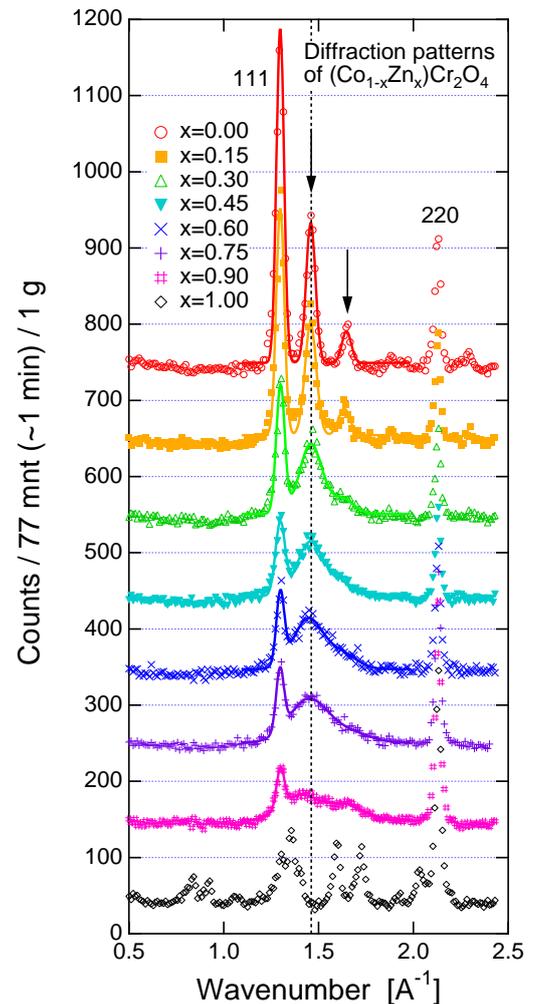


Fig. 1. Elastic scattering data of  $(\text{Co}_{1-x}\text{Zn}_x)\text{Cr}_2\text{O}_4$  in lowest temperature phases.