Supplemental Information

Phase transitions and slow spin dynamics of slightly inverted A-site spinel $CoAl_{2-x}Ga_xO_4$

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Crystallographic analysis

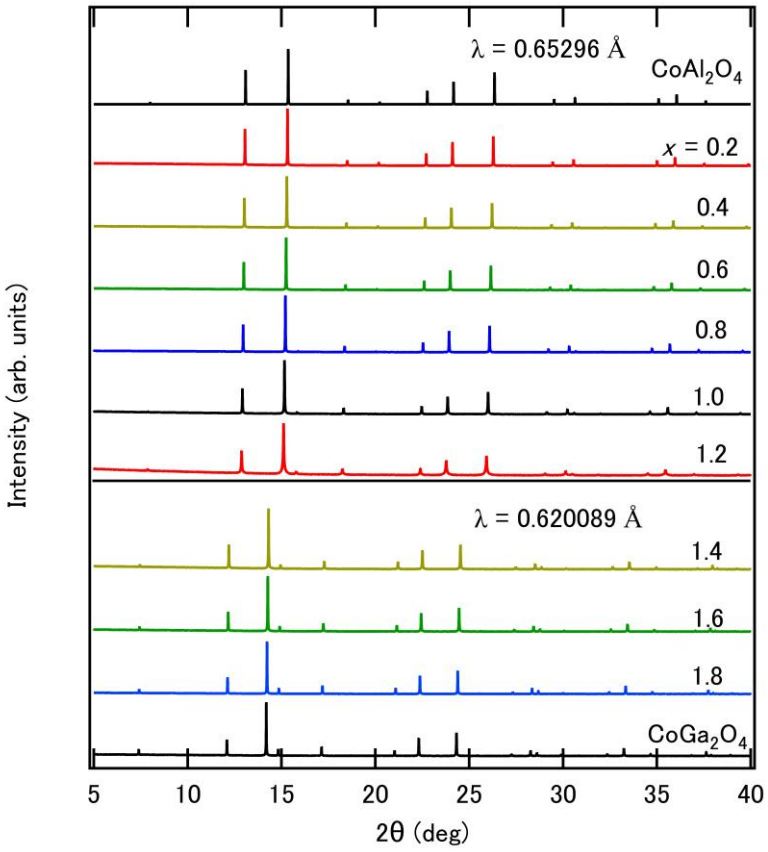

FIG. S1. XRD profiles for $CoAl_{2-x}Ga_xO_4$ obtained with a monochromatic synchrotron x-ray source with wavelengths of 0.65298 Å for $0 \le x \le 1.2$ and 0.620089 Å for $1.4 \le x \le 2.0$.

Table S-I. Crystallographic parameters for $CoAl_{2-x}Ga_xO_4$ refined with the Rietveld method.[a]

x	a (Å)	u	g_A(Co)	g_A(Al)	g_A(Ga)	R_{wp}(%)	S
0	8.10301(6)	0.2646(1)	0.945(3)	0.055(3)	-	-	-
0.2	8.12408(1)	0.26340(4)	0.847(3)	0.030(3)	0.123(9)	2.01	2.60
0.3	8.13424(1)	0.26307(5)	0.817(5)	0.017(5)	0.166(16)	2.71	2.23
0.4	8.14415(1)	0.26247(5)	0.795(4)	0.000(5)	0.204(19)	2.20	3.04
0.6	8.16405(1)	0.26130(6)	0.658(4)	0	0.342(4)	2.18	2.94
0.8	8.18500(1)	0.26058(6)	0.557(4)	0	0.443(4)	2.37	3.38
1.0	8.20969(1)	0.2599(1)	0.501(6)	0	0.499(6)	3.57	2.66
1.2	8.23611(5)	0.2593(1)	0.388(12)	0.028(12)	0.584(45)	3.71	2.77
1.4	8.25618(2)	0.2593(1)	0.381(7)	0	0.619(7)	3.20	2.94
1.6	8.28018(2)	0.2592(1)	0.393(6)	0	0.607(6)	3.47	2.54
1.8	8.30383(1)	0.2590(1)	0.377(6)	0	0.623(6)	3.74	3.39
2.0	8.32654(1)	0.2584(1)	0.336(7)	0	0.664(7)	4.04	3.56

[a]The cationic configuration was refined with the confinements $g_A(Co) = 1 - g_A(Al) - g_A(Ga)$, $g_B(Co) = [g_A(Al) + g_A(Ga)]/2$, $g_B(Al) = 1 - [x + g_A(Al)]/2$, and $g_B(Ga) = [x - g_A(Ga)]/2$. Note that, $\eta = 1 - g_A(Co)$. Data points for $x = 0$ and $= 2.0$ are from previous studies [9] and [5], respectively.

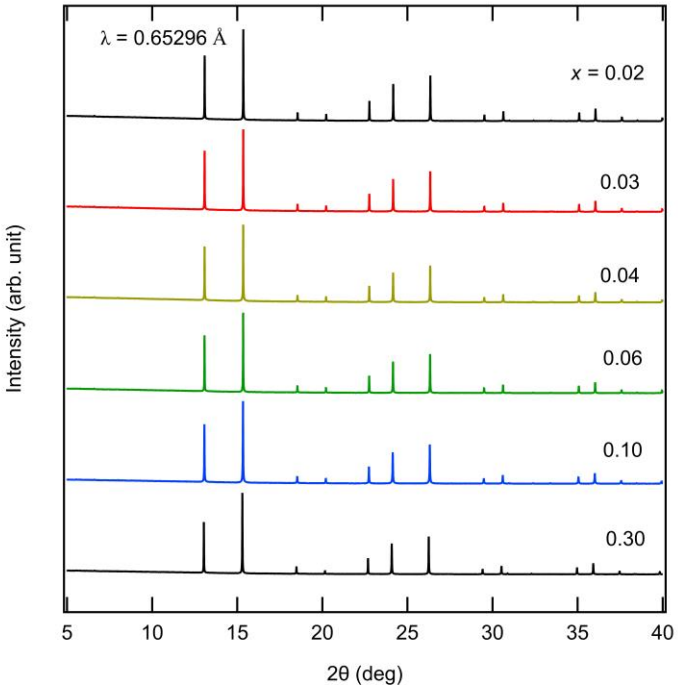

FIG. S2. XRD profiles for $CoAl_{2-x}Ga_xO_4$ obtained with a monochromatic synchrotron x-ray source with a wavelength of 0.65296 Å for $0.02 \leq x \leq 0.30$.

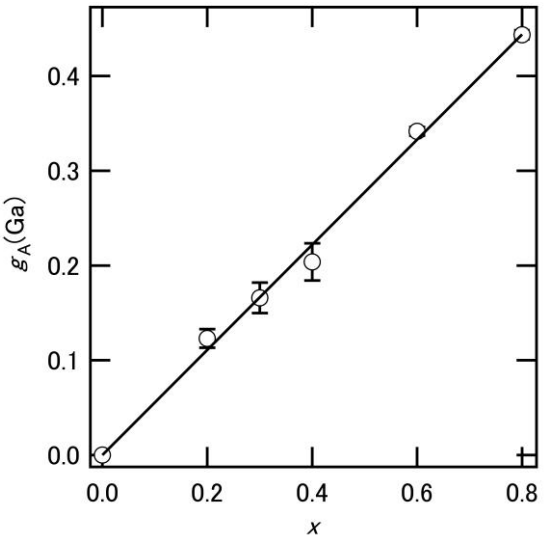

Fig. S3. A-site occupancy of Ga ions $g_A(Ga)$ for $x < 0.8$. Solid line corresponds to $g_A(Ga) = mx$. The constant $m = 0.56(1)$ was obtained by least squares.

Table S-II. Crystallographic parameters for $CoAl_{2-x}Ga_xO_4$ in the range $0.01 \le x \le 0.10$, refined with the Rietveld method.[b]

x	a (Å)	u	g_A(Co)	g_A(Al)	R_{wp}(%)	S
0.02	8.10561(1)	0.26443(4)	0.936(1)	0.053(1)	2.58	1.94
0.03	8.10720(1)	0.26438(5)	0.932(1)	0.051(1)	3.02	2.19
0.04	8.10802(1)	0.26432(6)	0.928(1)	0.050(1)	2.19	2.39
0.06	8.11025(1)	0.26425(5)	0.920(1)	0.046(1)	2.77	2.10
0.10	8.11479(1)	0.26411(5)	0.902(1)	0.042(1)	3.19	2.56

[b]The x-variation of g_A(Ga) was assumed to be $g_A(Ga) = 0.56x$ in the range $0.02 \le x \le 0.10$ (See Fig. S3).

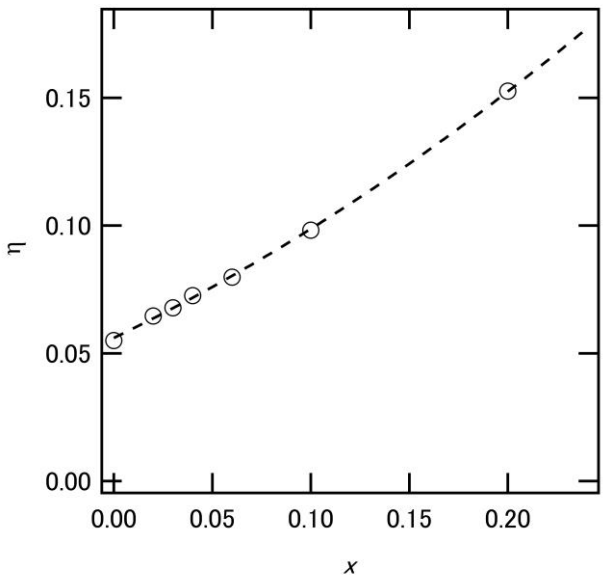

Fig. S4. Inversion parameter η as a function of x (for $x \le 0.2$) obtained on the assumption that $g_A(Ga) = 0.56x$. Dashed line represents the polynomial curve $\eta(x) = K_0 + K_1x + K_2x^2$ that was obtained by least squares, with $K_0 = 0.0560(7)$, $K_1 = 0.37(2)$, and $K_2 = 0.55(9)$. From the polynomial of $\eta(x)$, we determined that $\eta = 0.060(2)$, 0.076(1), and 0.089(2) for $x = 0.01$, 0.05, and 0.08, respectively.