Supporting Information for

Rational design, crystal structure, and frustrated magnetism of the Ge-containing $YbFe_2O_4$-type layered oxides $In_2Zn_{3-x}Co_xGeO_8$ ($0 \leq x \leq 3$)

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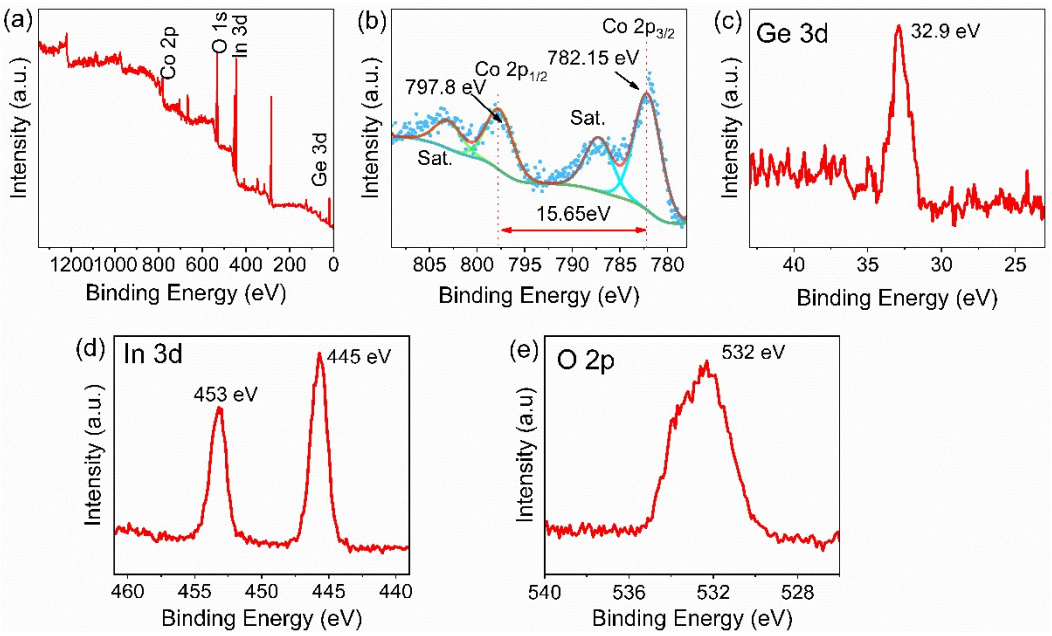

Fig. S1 (a) The survey XPS of $In_2Co_3GeO_8$. XPS spectra of Co 2p (b), Ge 3d (c), In 3d (d), and O 2p (e).

Table S1. Crystallographic data for $In_3Zn_{4.5}Ge_{1.5}O_{12}$, $In_3Zn_3Co_{1.5}Ge_{1.5}O_{12}$, $In_3Zn_{1.5}Co_3Ge_{1.5}O_{12}$ and $In_3Co_{4.5}Ge_{1.5}O_{12}$, corresponding to $In_2Zn_{3-x}Co_xGeO_8$ with x = 0, 1, 2, and 3, respectively.

parameters				
temperature (K)	300			
formula	$In_3Zn_{4.5}Ge_{1.5}O_{12}$	$In_3Zn_3Co_{1.5}Ge_{1.5}O_{12}$	$In_3Zn_{1.5}Co_3Ge_{1.5}O_{12}$	$In_3Co_{4.5}Ge_{1.5}O_{12}$
formula weight (g mol^{-1})	939.747	930.034	920.320	910.606
crystal system	Trigonal			
Space group (No.)	*R*-3*m* (166)			
wavelength (Å)	1.54059			
d spacing range	0.82−8.8	0.82-8.8	0.82−8.8	0.82−8.8
density (g cm^{-3})	6.300(4)	6.216(3)	6.154(4)	6.113(6)
unit cell dimensions	*a* = *b* = 3.2989(1) *c* = 26.3198(7)	*a* = *b* = 3.30826(7) *c* = 26.2143(6)	*a* = *b* = 3.31488(6) *c* = 26.0974(5)	*a* = *b* = 3.3159 (1) *c* = 25.9718(5)
V	248.06(2)	248.47(1)	248.47(1)	247.30(2)
Z	1	1	1	1
no. of reflections	85	85	85	85
no. of parameters	36	42	42	36
no. of constrains	6	10	10	6
R_{wp}	7.95%	2.68%	2.12%	2.23
R_p	5.89%	1.94%	1.26%	1.75
goodness of fit	1.78	1.40	2.36	1.12

Table S2. Atomic coordinates, site occupancy factors (*sof.*), and isotropic thermal displacement factors for $In_2Zn_{3-x}Co_xGeO_8$ (x = 1 and 2) obtained from Rietveld refinements against PXRD data.

***x* = 1**	Site	*x*	*y*	*z*	*sof.*	$B_{iso.}$ (Å^2)
In1	3a	0	0	0	0.897(1)	0.14(1)
Zn1	3a	0	0	0	0.069(1)	0.14(1)
Co1	3a	0	0	0	0.034(1)	0.14(1)
Zn2	6c	2/3	1/3	0.11563(2)	0.4656(5)	0.82(2)
In2	6c	2/3	1/3	0.11563(2)	0.0516(7)	0.82(2)
Co2	6c	2/3	1/3	0.11563(2)	0.2328(2)	0.82(2)
Ge1	6c	2/3	1/3	0.11563(2)	0.25	0.72(5)
O1	6c	0	0	0.13035(9)	1	1.30(8)
O2	6c	2/3	1/3	0.0388(1)	1	0.47(6)
***x* = 2**						
In1	3a	0	0	0	0.935(2)	0.27(2)
Zn1	3a	0	0	0	0.022(1)	0.27(2)
Co1	3a	0	0	0	0.043(2)	0.27(2)
Co2	6c	2/3	1/3	0.11598(3)	0.4784(5)	0.58(3)
In2	6c	2/3	1/3	0.11598(3)	0.0326(8)	0.58(3)
Zn2	6c	2/3	1/3	0.11598(3)	0.2390(3)	0.58(3)
Ge1	6c	2/3	1/3	0.11598(3)	0.25	0.58(3)
O1	6c	0	0	0.1308(1)	1	1.3(1)
O2	6c	2/3	1/3	0.0403(2)	1	0.30(9)

Table S3. Selected interatomic bond lengths of $In_2Zn_{3-x}Co_xGeO_8$ ($0 \leq x \leq 3$).

$In_2Zn_3GeO_8$			
In-O2 (×6)	2.170(2)	*M*-O2	1.997(3)
M-O1 (×3)	1.9358(6)	*M*-O1	2.356(3)
$In_2Zn_2CoGeO_8$			
In-O2 (×6)	2.164(2)	*M*-O2	2.014(3)
M-O1 (×3)	1.9486(5)	*M*-O1	2.290(2)
$In_2ZnCo_2GeO_8$			
In-O2 (×6)	2.184(2)	*M*-O2	1.975(4)
M-O1 (×3)	1.9523(7)	*M*-O1	2.260(3)
$In_2Co_3GeO_8$			
In-O2 (×6)	2.205(4)	*M*-O2	1.917(8)
M-O1 (×3)	1.964(1)	*M*-O1	2.194(5)