Supporting Information

**Synthesis and Structure of Four $\text{Sc}_3\text{Cu}_y\text{Zn}_{18-y}$ -Type Compositions ($0 \leq y \leq \sim 2.2$). A
1/1 Crystalline Approximants of a New Icosahedral Quasicrystal. Direct Example of
Tuning on the Basis of Size Effects and Hume-Rothery Concepts**

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TABLE S1. Data collection and refinement parameters for crystalline products of the nominal $\text{Sc}_3\text{Cu}_x\text{Zn}_{17-x}$ reactions.

Reactant x	0	1	2	3
empir. form. found	$(\text{ScZn}_6)_3$	$\text{Sc}_3\text{Cu}_{0.65}\text{Zn}_{17.4}$	$\text{Sc}_3\text{Cu}_{1.486}\text{Zn}_{16.5}$	$\text{Sc}_3\text{Cu}_{2.19}\text{Zn}_{16.0}$
space group, Z	$Im\bar{3}, 8$	$Im\bar{3}, 8$	$Im\bar{3}, 8$	$Im\bar{3}, 8$
latt. para. ^a				
a (Å)	13.8311(5)	13.7923(5)	13.7598(6)	13.7528(5)
V (Å ³)	2645.9(2)	2623.7(2)	2605.2(2)	2601.2(2)
d_{calc} (g/cm ³)	6.554	6.635	6.674	6.785
cryst. dim. (mm)	$0.08 \times 0.12 \times 0.13$	$0.08 \times 0.10 \times 0.16$	$0.16 \times 0.20 \times 0.25$	$0.15 \times 0.20 \times 0.25$
diffractometer	Siemens APEX	Siemens APEX	Siemens APEX	Siemens APEX
radiation; $2\theta_{\text{max}}$	Mo $K\alpha$, 56.56	Mo $K\alpha$, 56.56	Mo $K\alpha$, 56.56	Mo $K\alpha$, 56.56
index ranges	$-17 \leq h, k \leq 18;$ $-14 \leq l \leq 18$	$-18 \leq h \leq 13;$ $-18 \leq k \leq 17;$ $-18 \leq l \leq 12$	$-18 \leq h \leq 16;$ $-17 \leq k \leq 18;$ $-18 \leq l \leq 15$	$-11 \leq h \leq 17;$ $-17 \leq k \leq 18;$ $-18 \leq l \leq 18$
μ (mm ⁻¹)(Mo $K\alpha$)	33.14	33.46	33.52	33.99
no. of refl.				
measured	8387	8274	8185	8141
unique obs. ($>2\sigma_I$)	608	604	600	600
no. of variables	47	47	47	50

$R_{\text{avg}} (I \geq 2\sigma_I)(\%)$	4.25	4.21	3.81	4.43
$R1, wR2 (\%)$	2.77, 6.02	2.38, 4.77	2.87, 5.52	3.98, 8.05
goodness of fit	1.06	1.13	1.16	1.09
$\Delta F_{\text{max}} (e^-/\text{\AA}^3)$	5.91 (1.52\text{\AA} from Zn7)	1.76 (0.74\text{\AA} from Zn7)	3.12 (2.08\text{\AA} from Zn6)	5.67 (0.88\text{\AA} from Zn6)
$\Delta F_{\text{min}} (e^-/\text{\AA}^3)$	-1.59 (1.14\text{\AA} from Zn7)	-1.52 (0.75\text{\AA} from Zn5)	-1.93 (0.62\text{\AA} from Zn7)	-2.69 (0.62\text{\AA} from Zn6)
Extinct. coeff. (10^{-6})	6(2)	59(2)	136(15)	21(2)

^a Huber diffractometer data, $\lambda = 1.540598 \text{ \AA}$, 23 °C.

Table S2. Positional ($\times 10^4$) and Displacement Parameters ($\times 10^3$) for $\text{Sc}_3\text{Cu}_y\text{Zn}_{18-y}$ ($y = 0 - 2.12$).^a

Atom	Wyckoff	Site symm	<i>x</i>	<i>y</i>	<i>z</i>	<i>U</i> (eq) ^b	Occupancy:Zn/Cu
Zn1	12 <i>e</i>	mm2..	1901(1)	0	½	9(1)	
			1900(1)	0	½	10(1)	
			1895(1)	0	½	8(1)	
			1887(1)	0	½	8(1)	
Zn2	24 <i>g</i>	m..	0	4044(1)	3457(1)	8(1)	
			0	4041(1)	3452(1)	9(1)	
			0	4045(1)	3449(1)	6(1)	
			0	4051(1)	3448(1)	6(1)	
Zn3	48 <i>h</i>	1	1169(1)	3407(1)	1998(1)	12(1)	
			1157(1)	3418(1)	2004(1)	12(1)	
			1144(1)	3426(1)	2001(1)	8(1)	
			1125(1)	3437(1)	1992(1)	10(1)	
Zn4	12 <i>d</i>	mm2..	4053(1)	0	0	20(1)	
			4073(1)	0	0	21(1)	
			4079(1)	0	0	15(1)	
			4079(1)	0	0	13(1)	
Zn5/Cu	24 <i>g</i>	m..	0	2382(1)	916(1)	24(1)	100/0
			0	2390(1)	917(1)	24(1)	78/22
			0	2388(1)	912(1)	21(1)	88/12
			0	2396(1)	906(1)	18(1)	82/18

Zn6	16f	.3.	1617(1)	1617(1)	1617(1)	17(1)	
			1626(1)	1626(1)	1626(1)	16(1)	
			1626(1)	1626(1)	1626(1)	16(1)	
			1621(1)	1621(1)	1621(1)	25(1)	
Zn7	24g	m..	0	748(6)	810(5)	99(3)	0.300(5)/0
			0	787(4)	788(4)	89(2)	0.334(5)/0
			0	760(5)	823(5)	88(3)	0.333(6)/0
			0	765(9)	782(8)	130(6)	0.349(9)/0
Sc	24g	m..	0	1898(1)	2997(1)	5(1)	
			0	1882(1)	3002(1)	7(1)	
			0	1874(1)	3001(1)	4(1)	
				1873(1)	2995(1)	5(1)	
Zn8	8c	.3.	-	-	-	-	-
			-	-	-	-	-
			-	-	-	-	-
			1/4	1/4	1/4	75(10)	24(2)%

^a Parameters are listed in sequence for ScZn₆, Sc₃Cu_{0.65}Zn_{17.4}, Sc₃Cu_{1.48}Zn_{16.5}, and Sc₃Cu_{2.19}Zn_{16.0}, respectively.

^b $U(\text{eq})$ is defined as one third of the trace of the orthogonalized U_{ij} tensor.

^c Mixed occupancy was fixed at EDX proportion for the ternary crystals, while occupancy at Zn7 position was fixed at 1/3 in analyses in article. The refined occupancies of Zn7 were as listed above.

Table S3. Anisotropic Displacement Parameters ($\times 10^3$) for $\text{Sc}_3\text{Cu}_y\text{Zn}_{18-y}$ ($y = 0 - 2.12$)^a

Atom	U_{11}^b	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Zn1	7(1)	7(1)	15(1)	0	0	0
	9(1)	8(1)	15(1)	0	0	0
	6(1)	5(1)	12(1)	0	0	0
	6(1)	5(1)	12(1)	0	0	0
Zn2	9(1)	6(1)	7(1)	2(1)	0	0
	10(1)	8(1)	8(1)	2(1)	0	0
	7(1)	6(1)	6(1)	2(1)	0	0
	7(1)	6(1)	6(1)	1(1)	0	0
Zn3	17(1)	9(1)	10(1)	-1(1)	6(1)	-3(1)
	16(1)	9(1)	11(1)	-1(1)	5(1)	-3(1)
	13(1)	6(1)	8(1)	-1(1)	5(1)	-3(1)
	14(1)	7(1)	9(1)	-2(1)	5(1)	-3(1)
Zn4	10(1)	39(1)	10(1)	0	0	0
	10(1)	41(1)	11(1)	0	0	0
	6(1)	32(1)	8(1)	0	0	0
	6(1)	24(1)	8(1)	0	0	0
Zn5/Cu	9(1)	54(1)	9(1)	10(1)	0	0
	9(1)	53(1)	10(1)	11(1)	0	0
	6(1)	49(1)	7(1)	11(1)	0	0
	7(1)	41(1)	7(1)	11(1)	0	0

Zn6	17(1)	17(1)	17(1)	11(1)	11(1)	11(1)
	16(1)	16(1)	16(1)	8(1)	8(1)	8(1)
	16(1)	16(1)	16(1)	10(1)	10(1)	10(1)
	25(1)	25(1)	25(1)	17(1)	17(1)	17(1)
Zn7	190(7)	59(4)	47(4)	-34(3)	0	0
	167(5)	53(3)	45(3)	-33(2)	0	0
	174(6)	45(3)	45(4)	-33(3)	0	0
	279(14)	61(7)	50(6)	-40(4)	0	0
Sc	6(1)	6(1)	7(1)	0	0	0
	5(1)	7(1)	8(1)	1(1)	0	0
	3(1)	5(1)	5(1)	1(1)	0	0
	3(1)	5(1)	6(1)	1(1)	0	0
Zn8	-	-	-	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-
	75(10)	75(10)	75(10)	66(10)	66(10)	66(10)

^a Parameters are listed in sequence for ScZn₆, Sc₃Cu_{0.65}Zn_{17.4}, Sc₃Cu_{1.48}Zn_{16.5}, and Sc₃Cu_{2.19}Zn_{16.0}, respectively.

^b General expression of the thermal parameter is $\exp -2\pi^2 [U_{11}h^2a^{*2} + U_{22}k^2b^{*2} + U_{33}l^2c^{*2} + 2U_{12}hka^*b^*c^* + 2U_{13}hla^*c^* + U_{23}klb^*c^*]$

Table S4. Bond distance in $\text{Sc}_3\text{Cu}_x\text{Zn}_{18-x}$

Bonds	$\text{Sc}_3\text{Zn}_{18}$	$\text{Sc}_3\text{Cu}_{0.65}\text{Zn}_{17.4}$	$\text{Sc}_3\text{Cu}_{1.48}\text{Zn}_{16.5}$	$\text{Sc}_3\text{Cu}_{2.19}\text{Zn}_{16.0}$	Bonds	$\text{Sc}_3\text{Zn}_{18}$	$\text{Sc}_3\text{Cu}_{0.65}\text{Zn}_{17.4}$	$\text{Sc}_3\text{Cu}_{1.48}\text{Zn}_{16.5}$	$\text{Sc}_3\text{Cu}_{2.19}\text{Zn}_{16.0}$
Zn1	Zn2	2x	2.502(1)	2.4990(9)					
	Zn2	2x	2.526(1)	2.516(1)					
	Zn3	4x	2.7360(6)	2.7072(6)					
	Zn4	2x	2.938(1)	2.916(1)					
	Sc	2x	3.231(1)	3.228(1)					
Zn2	Zn1	1x	2.502(1)	2.4990(9)					
	Zn1	1x	2.526(1)	2.516(1)					
	Zn2	1x	2.645(2)	2.645(2)					
	Zn3	2x	2.6824(8)	2.6705(7)					
	Zn3	2x	2.7321(9)	2.6967(8)					
	Zn4	2x	2.832(1)	2.8186(9)					
	Sc	1x	3.035(2)	3.043(1)					
	Sc	2x	3.109(1)	3.091(1)					
Zn3	Zn6	1x	2.6060(9)	2.6076(8)					
	Zn5	1x	2.620(1)	2.608(1)					
	Zn3	2x	2.6621(9)	2.6675(8)					
	Zn2	1x	2.6824(8)	2.6705(7)					
	Zn2	1x	2.7321(9)	2.6967(8)					
	Zn1	1x	2.7360(6)	2.7072(6)					
	Sc	1x	2.919(1)	2.9104(9)					
	Sc	1x	2.980(1)	2.988(1)					

	Zn5								
	Zn7	1x	2.264(8)	2.217(5)					
	Zn5	1x	2.534(2)	2.530(2)					
	Zn3	2x	2.620(1)	2.608(1)					
	Zn4	1x	2.636(2)	2.643(2)					
	Zn6	2x	2.6572(8)	2.6636(7)					
	Zn7	2x	2.720(8)	2.770(6)					
	Sc	1x	2.955(2)	2.959(1)					
	Sc	2x	3.037(1)	3.008(1)					
	Zn3	3x	2.6060(9)	2.608(1)					
	Zn5	3x	2.6572(8)	2.6636(7)					
	Zn7	3x	2.774(3)	2.775(3)					
	Sc	3x	2.9656(9)	2.9588(8)					
	Zn3	2x	2.919(1)	2.9104(9)					
	Zn5	1x	2.955(2)	2.959(1)					
	Zn6	2x	2.9656(9)	2.9588(8)					
	Zn3	2x	2.980(1)	2.988(1)					
	Zn3	2x	2.9954(8)	2.9946(7)					
	Zn4	1x	3.004(1)	2.986(1)					
	Zn2	1x	3.035(2)	3.043(1)					
	Zn5	2x	3.037(1)	3.008(1)					
	Zn2	2x	3.109(1)	3.091(1)					

	Sc	1x	2.9954(8)	2.9946(7)	2.9878(8)	2.989(1)	Zn7	Zn5	1x	2.264(8)	2.217(5)	2.260(1)	2.25(1)
Zn4	Zn4	1x	2.621(3)	2.557(2)	2.534(3)	2.533(4)		Zn5	2x	2.720(8)	2.770(6)	2.766(3)	2.754(1)
	Zn5	2x	2.636(2)	2.643(2)	2.644(2)	2.629(2)		Zn6	2x	2.774(3)	2.775(3)	2.805(7)	2.772(5)
	Zn2	4x	2.832(1)	2.8186(9)	2.809(1)	2.804(1)	Zn8	Zn6	2x				2.094(2)
	Zn1	2x	2.938(1)	2.916(1)	2.899(1)	2.887(2)		Zn3	6x				2.393(1)
	Sc	2x	3.004(1)	2.986(1)	2.975(2)	2.977(2)

Fig. S1. Detailed comparison of experimental powder patterns of nominal $\text{Sc}_3\text{Zn}_{17}$ and $\text{Sc}_3\text{Zn}_{18}$.