Supporting Information for “Valence Instabilities, Phase Transitions and Abrupt Lattice Expansion at 5 K in the YbGaGe System”

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Figure 1S. Final observed (o) and calculated (solid line) powder synchrotron X-ray ($\lambda = 0.42970 \text{ \AA}$) diffraction profiles for YbGaSn at 300 K. The lower solid line shows the difference profile and the tick marks show the reflection positions.

Figure 2S. Temperature evolution of (a) the hexagonal lattice constants and the unit cell volume and (b) selected interatomic distances in YbGaSn.

Figure 3S. Final observed (o) and calculated (solid line) powder synchrotron X-ray ($\lambda = 0.42970$ Å) diffraction profiles for $\text{YbGa}_{1.05}\text{Ge}_{0.95}$ at 5 K. The lower solid line shows the difference profile and the tick marks show the reflection positions of the two co-existing hexagonal phases.

Figure 4S. Selected regions of the diffraction profile of $\text{YbGa}_{1.05}\text{Ge}_{0.95}$ showing the temperature evolution of the (100) and (104) Bragg reflections ($\lambda = 0.42970 \text{ \AA}$) at 673, 823 and 923 K.

Figure 5S. Final observed (o) and calculated (solid line) powder synchrotron X-ray ($\lambda = 0.42970$ Å) diffraction profiles for $\text{YbGa}_{1.05}\text{Ge}_{0.95}$ at 1123 K. The lower solid line shows the difference profile and the tick marks show the reflection positions.

Table 1S. Refined parameters for YbGaSn as obtained by Rietveld refinement of the synchrotron X-ray powder diffraction data at 300 K in the space group $P6_3/mmc$ with unit cell parameters, $a = 4.44535(3)$ Å and $c = 17.3245(2)$ Å (agreement factors: $R_{wp} = 8.63\%$, $R_{exp} = 1.01\%$, $R_{F2} = 11.93\%$).

atom	site	x/a	y/b	z/c	$B_{iso}/\text{\AA}^2$	n
Yb ⁺² (1)	$2b$	0	0	1/4	0.67(3)	1.088(4)
Yb ⁺² (2)	$2a$	0	0	0	1.32(3)	1.066(4)
Ga	$4f$	1/3	2/3	0.1640(2)	0.64(5)	1.099(7)
Sn	$4f$	1/3	2/3	0.6152(2)	0.80(5)	0.972(4)

Table 2S. Refined parameters for YbGa_{1.05}Ge_{0.95} as obtained by Rietveld refinement of the synchrotron X-ray powder diffraction data at 305 K in the space group $P6_3/mmc$ with unit cell parameters, $a = 4.211170(8)$ Å and $c = 16.80480(7)$ Å ($R_{wp} = 7.09\%$, $R_{exp} = 1.21\%$, $R_{F2} = 5.63\%$).

atom	site	x/a	y/b	z/c	$B_{iso}/\text{\AA}^2$	n
Yb ⁺³ (1)	$2b$	0	0	1/4	1.09(5)	1.010(3)
Yb ⁺² (2)	$2a$	0	0	0	0.49(5)	0.989(3)
Ga	$4f$	1/3	2/3	0.1518(1)	0.85(6)	0.995(3)
Ge	$4f$	1/3	2/3	0.6125(1)	0.53(4)	0.953(3)
Ga'	$4f$	1/3	2/3	0.6125(1)	0.53(4)	0.056(3)

Table 3S. Refined parameters for YbGa_{1.05}Ge_{0.95} as obtained by Rietveld refinement of the synchrotron X-ray powder diffraction data at 5 K (R_{wp} = 9.67%, R_{exp} = 1.76%, R_{F2} = 7.85%).

<i>Phase I</i> space group $P6_3/mmc$ $a = 4.22000(1) \text{ \AA}$, $c = 16.88466(9) \text{ \AA}$ phase fraction 62.0(2)%						
atom	site	x/a	y/b	z/c	$B_{iso}/\text{\AA}^2$	n
Yb ⁺² (1)	2 <i>b</i>	0	0	1/4	0.91(2)	1.04(1)
Yb ⁺² (2)	2 <i>a</i>	0	0	0	0.52(2)	1.00(1)
Ga	4 <i>f</i>	1/3	2/3	0.1498(2)	1.36(8)	0.995
Ge	4 <i>f</i>	1/3	2/3	0.6102(3)	0.18(4)	0.953
Ga	4 <i>f</i>	1/3	2/3	0.6102(3)	0.18(4)	0.056
<i>Phase II</i> space group $P6_3/mmc$ $a = 4.20169(1) \text{ \AA}$, $c = 16.7175(1) \text{ \AA}$ phase fraction 38.0(4)%						
atom	site	x/a	y/b	z/c	$B_{iso}/\text{\AA}^2$	n
Yb ⁺³ (1)	2 <i>b</i>	0	0	1/4	0.45(3)	1.02(1)
Yb ⁺² (2)	2 <i>a</i>	0	0	0	0.52(3)	0.99(1)
Ga	4 <i>f</i>	1/3	2/3	0.1539(3)	0.33(8)	0.995
Ge	4 <i>f</i>	1/3	2/3	0.6120(4)	0.11(4)	0.953
Ga	4 <i>f</i>	1/3	2/3	0.6120(4)	0.11(4)	0.056

Table 4S. Refined parameters for YbGa_{1.05}Ge_{0.95} as obtained by Rietveld refinement of the synchrotron X-ray powder diffraction data at 1123 K in the space group *P6/mmm* with unit cell parameters, $a = 4.231908(3)$ Å and $c = 4.342763(4)$ Å ($R_{\text{wp}} = 8.35\%$, $R_{\text{exp}} = 3.28\%$, $R_{\text{F2}} = 5.55\%$).

atom	site	x/a	y/b	z/c	$B_{\text{iso}}/\text{\AA}^2$	n
Yb ⁺³	$1a$	0	0	0	2.99(2)	1.196(1)
Ga	$2d$	1/3	2/3	1/2	3.72(4)	0.551(2)
Ge	$2d$	1/3	2/3	1/2	3.65(4)	0.486(2)