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In Situ Neutron Powder Diffraction of the Formation of $SrGa_2D_2$, and Hydrogenation Behavior of $YbGa_2$ and $EuGa_2$

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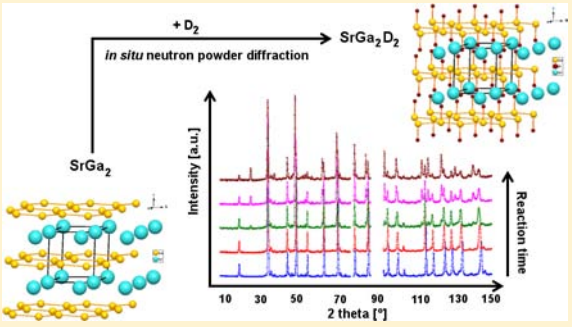
ABSTRACT: The hydrogenation behavior of the Zintl phases MGa_2 (M = Sr, Eu, Yb) was investigated. Upon being heated under 50 bar of hydrogen gas pressure, $SrGa_2$ starts forming the polyanionic hydride $SrGa_2H_2$ at 125 °C as observed by in situ differential scanning calorimetry (DSC) and ex situ X-ray powder diffraction. The deuteration of $SrGa_2$ was studied by in situ neutron powder diffraction at temperatures up to 300 °C in the pressure range of $50 \leq p(D_2) \leq 61$ bar with a time resolution of two minutes. Neither incorporation of deuterium into the interstitials of the crystal structure of $SrGa_2$ nor any other intermediate of the reaction was observed. No significant variation in positional or occupational parameters as refined by the Rietveld method was observed for the deuteride $SrGa_2D_2$ under various temperature and pressure conditions (e.g., at T = 299 °C, $p(D_2)$ = 60 bar, space group $P\bar{3}m1$, a = 4.4098(2) Å, c = 4.7429(3) Å, Sr in 1*a*, 0 0 0; Ga in 2*d*, 1/3 2/3 0.474(1); D in 2*d*, 1/3 2/3 0.120(2)). In situ DSC complemented by ex situ X-ray powder diffraction revealed that $EuGa_2$ and $YbGa_2$ do not form polyanionic hydrides in the investigated temperature−pressure ranges (20 °C ≤ T ≤ 220 °C, 5 bar ≤ p ≤ 54 bar). Instead binary hydrides and gallium-rich intermetallics are formed. The first refined crystal structural data are presented for $YbGa_6$ in the tetragonal $PuGa_6$-type structure (space group $P4/nbm$ (No. 125), a = 6.0890(3) Å, c = 7.6562(4) Å, Yb in 2*c*, 3/4 1/4 0; Ga1 in 8*m*, 0.4750(4) 0.5250(4) 0.3377(4); Ga2 in 4*g*, 1/4 1/4 0.1691(5)).



1. INTRODUCTION

In this paper, results of in situ neutron powder diffraction (NPD) experiments on the Zintl phase $SrGa_2$ and its deuteride are presented. Also the related phases $YbGa_2$ and $EuGa_2$ were investigated with respect to a possible hydride formation.

All three compounds are Zintl phases structurally related to each other. $SrGa_2$ crystallizes in the hexagonal AlB_2-type, $YbGa_2$ in the hexagonal $CaIn_2$-type,[1,2] and $EuGa_2$ in the orthorhombic KHg_2-type, which is very similar to the $CeCu_2$-type.[1,3] In all of these structures, Ga forms graphite-like hexagonal layers, which are more or less distorted and connected or isolated. The Sr-, Yb-, or Eu-atoms are situated between the Ga layers (Figure 1). The structural relation can also be shown by the use of a Bärnighausen tree.[4]

Hydrides from Zintl phases can be categorized after Häussermann[5] as either "Zintl phase hydrides" (e.g., CaSi−$CaSiH_{1.0-1.3}$) or "polyanionic hydrides" (e.g., $SrAl_2$−$SrAl_2H_2$). While the former contain hydride anions in interstitial positions, the latter are characterized by the presence of polyanions with homonuclear bonding to which the hydrogen atoms are covalently bound. Contrary to the hydrides with isolated $[EH_n]^{x-}$ units (e.g., borohydrides, alanates, and other), polyanionic hydrides are characterized by the presence of both E−E and E−H bonds.[5]

There are few other examples of polyanionic hydrides, such as MAlSiH (M = Ca, Sr, Ba), SrGaGeH, BaGaTtH (Tt = Si, Ge, Sn),[5] and $RbGaH_2$.[6] In contrast to the Zintl phase hydrides, all of these known polyanionic hydrides seem to be stoichiometric with respect to hydrogen. Therefore, we would like to know whether any intermediate phases, for example, hydrides with lower hydrogen content than the fully hydrogenated product, form during the formation of these hydrides.

Table 1. Experimental Conditions of in Situ DSC Experiments on the Hydrogenation of $YbGa_2$; Quantitative Phase Analysis by XRD of the Hydrogenated Products

hydrogen gas pressure [bar]	54	25	5
temperature[a] [°C]	220	180	180
phases/ weight fraction	$YbGa_6$/29.7(5)%	$YbGa_2$/25.7(6)%	$YbGa_4$/6.0(7)%
	$YbH_{2.67}$/70.3(5)%	$YbGa_4$/57(1)%	$YbGa_5$/14.4(2)%
		$YbGa_6$/11(1)%	$YbGa_6$/45.9(5)%
		$YbH_{2.67}$/5.2(6)%	$YbH_{2.67}$/33.7(5)%

[a]Held each time for 2 h.

For $SrGa_2$, the existence of a corresponding polyanionic hydride $SrGa_2H_2$ is already known. It forms at about 50 bar of hydrogen pressure and 180 °C.[7] Hydrides of the other compounds are not yet described in the literature, but because of the structural relationship, one can presume the existence of similar hydrides for $EuGa_2$ and $YbGa_2$.

2. EXPERIMENTAL SECTION

All samples were prepared from the metals. Gallium was obtained from ChemPur (pieces, 99.999%), ytterbium from Alfa Aesar (ingot, 99.9%),

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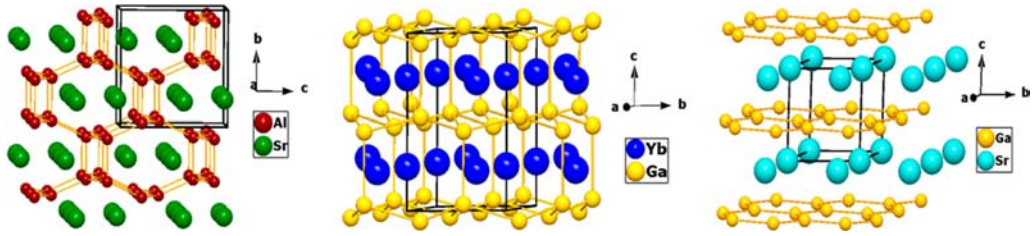

Figure 1. Structures of $SrAl_2$ ($CeCu_2$-type, left), $YbGa_2$ ($CaIn_2$-type, middle), and $SrGa_2$ (AlB_2-type, right).

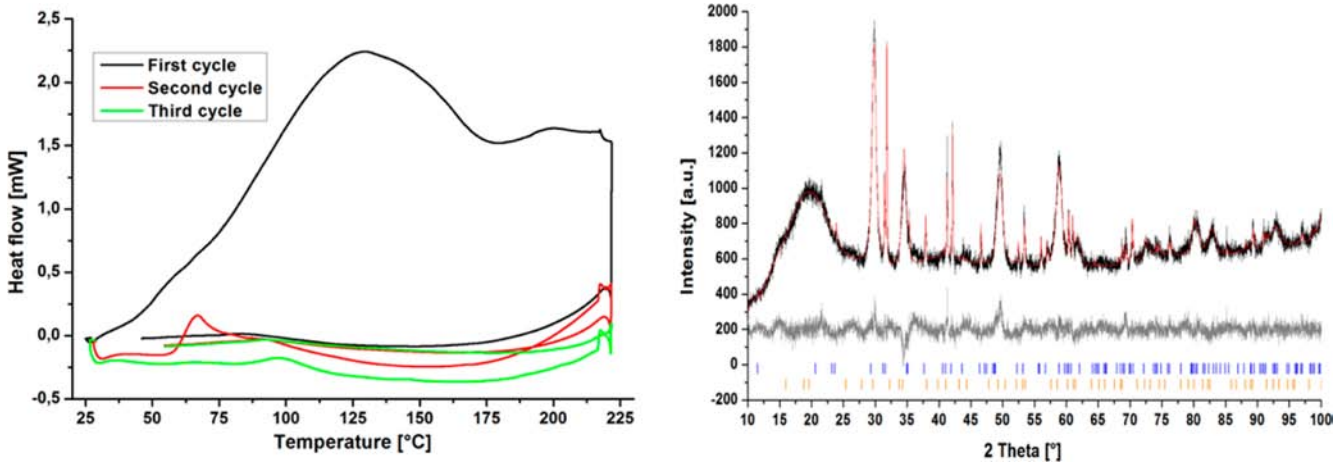

Figure 2. DSC diagram of $YbGa_2$ at 54 bar of hydrogen gas pressure and 220 °C for 2 h with exothermic signals in the positive direction (left) and X-ray powder pattern (Cu Kα) after the hydrogenation (right). Black is the measurement, red is the calculated curve, gray is the difference, and the Bragg markers from top to bottom are $YbGa_6$ and $YbH_{2.67}$.

Table 2. Refined $YbGa_6$ Crystallographic Data Prepared from DSC Experiments

		hydrogen pressure = 54 bar				hydrogen pressure = 5 bar			
space group		*P4/nbm* (No. 125)							
a [Å]		6.0921(3)				6.0890(3)			
c [Å]		7.6590(4)				7.6562(4)			
V [Å^3]		284.25(3)				283.86(3)			
		hydrogen pressure = 54 bar				hydrogen pressure = 5 bar			
site	Wyck. letter	*x*	*y*	*z*	B_{eq}	*x*	*y*	*z*	B_{eq}
Ga1	8m	0.477(1)	0.523(1)	0.3399(9)	1.0(2)	0.4750(4)	0.5250(4)	0.3377(4)	1.44(7)
Ga2	4g	1/4	1/4	0.166(1)	1.0(2)	1/4	1/4	0.1691(5)	1.44(7)
Yb	2c	3/4	1/4	0	1.0(2)	3/4	1/4	0	1.44(7)
occupancy		all sites 100%				all sites 100%			
R Bragg		1.93				1.57			
R_{wp}		4.79				2.55			

europium from Gerresheim Lübeck (ingot, 99.98%), and strontium from Alfa Aesar (distilled dendritic pieces, 99.8% metals basis). All handlings were performed in an argon-filled glovebox. The oxide surface layers of the metals were mechanically removed, and the metals were cut to pieces for the reaction. Stoichiometric amounts of the elements were mixed in welded Nb ampules and either sealed in silica ampules or heated in a sublimation tube under vacuum. The samples were heated to 800 or 850 °C (Eu) for about 30 h. The Zintl phases were obtained as brittle products of gray color with a slight metallic luster, which were easily ground to gray powders.

Hydrogenation experiments were carried out in a TA Instruments Q1000 differential scanning calorimeter (DSC) equipped with a gas pressure cell at various hydrogen gas pressures in crimped aluminum pans with sample amounts of approximately 10−30 mg. To investigate the reversibility of the hydrogenation reactions, the DSC experiments were, for each sample, repeated once or twice without opening the cell. The heating rate for all the experiments was 5 °C/min. Additionally, for $EuGa_2$ and $SrGa_2$, an autoclave machined from hydrogen-resistant Nicrofer 5219 Nb-alloy718 was used to prepare larger sample amounts. Hydrogen gas was obtained from Praxair (purity 99.9% or 99.999%).

Powder X-ray diffraction (XRD) data were taken with the use of a PANalytical XPert Pro diffractometer with PIXcel detector and Cu K$\alpha_{1/2}$ radiation in Bragg−Brentano geometry on flat samples enclosed between two kapton foils.

NPD experiments were carried out at the Institute Laue-Langevin in Grenoble, France, at high flux diffractometer D20 in high resolution mode with a time resolution of 2 min per pattern.

The sample was measured in a sapphire gas pressure cell[8] and heated with the use of a laser heating system (2 × 40 W). The deuterium (isotopic purity 99.8%) pressure was controlled with the use of a gas pressure regulation system. The temperature was measured with the use of a pyrometer (Keller HCW PL 11 AF 3). A calibration for the temperature was made with palladium lattice parameters with respect to the temperature dependence found by Dutta and Dayal[9] and given by eq 1.

$$T_{cal} = 8.0(7)\ ^\circ\mathrm{C} + 0.941(8)\cdot T_{Pyro} - 0.00029(2)\ ^\circ\mathrm{C}^{-1}\cdot T_{Pyro}^2 \quad (1)$$

For details of the experimental setup, see refs 8 and 10.

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Table 3. Experimental Conditions of in Situ DSC Experiments on the Hydrogenation of $EuGa_2$; Quantitative Phase Analysis by XRD of the Hydrogenated Products

pressure [bar]	50	25	1
temperature[a] [°C]	180	180	180
phases/ weight fraction	$EuGa_2$/36.2(7)%	$EuGa_2$/34.3(5)%	$EuGa_2$/19.7(4)%
	Eu_5Ga_9/12.5(7)%	Eu_5Ga_9/6.9(6)%	Eu_5Ga_9/5.6(4)%
	$EuGa_4$/28.8(6)%	$EuGa_4$/33.9(5)%	$EuGa_4$/41.0(5)%
	Eu_3Ga_8/22(1)%	Eu_3Ga_8/21.7(6)%	Eu_3Ga_8/30.3(5)%
	EuO/–	EuO/3.3(4)%	EuO/3.5(4)%

[a]Held each time for 2 h.

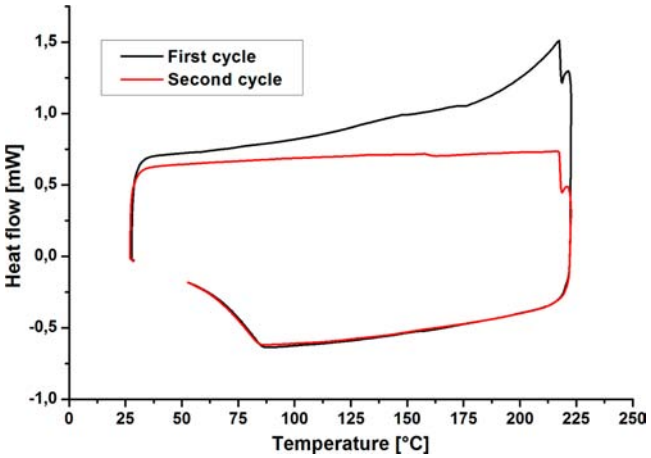

Figure 3. Heat flow curve of $SrGa_2$ at 50 bar H_2 pressure and 220 °C, held for 2 h, with exothermic signals in the positive direction.

The actual neutron wavelength, determined by a Rietveld refinement of a measurement of a NIST 640b silicon standard, was found to be 1.86765(5) Å. The Rietveld refinements of the XRD data were carried out with the programs TOPAS 2.1 and TOPAS 4.2,[11,12] and for the refinement of the NPD data the program FULLPROF[13] with pseudo-Voigt as the profile function was used.

3. RESULTS AND DISCUSSION

Hydrogenation Behavior of $YbGa_2$. The hydrogenation experiments were carried out with a single phase sample of $YbGa_2$ and with a sample of $YbGa_2$ containing about 4 wt % of YbGa as a secondary phase.

The DSC measurements show exothermic reactions with different H_2 pressures. The conditions were 54 bar and 220 °C for 2 h; 25 bar and 180 °C for 2 h; and 5 bar, 180 °C for 2 h. As shown in Figure 2 (left), for the measurement with 54 bar, significant heat effects in the first and second repetition are not reversible upon cooling down. This is typical for the formation of metal hydrides in such in situ DSC experiments.[14]

In all the cases, the XRD diffraction data of the gray hydrogenation products were taken. Other than Ga-rich phases such as $YbGa_4$, $YbGa_5$, and $YbGa_6$, the data show only the formation of $YbH_{2.67}$. $YbGa_5$ seems only to form at low hydrogen pressure. For details about the identified phases see Table 1. Note that, in all cases, gallium is missing with respect to the nominal composition, so the weight fractions do not give the exact composition. Missing gallium can be assumed to be amorphous, to have concentrated in grain boundaries, or to have interacted with the aluminum pan used. In Figure 2 (right) a Rietveld plot for the product of the hydrogenation at 54 bar is shown.

Tagawa et al. and Pelleg et al. identified $YbGa_6$ crystals in the $PuGa_6$-type structure and reported the lattice parameters, but did not specify the atom coordinates.[15,16] In our two DSC samples at 54 and 5 bar, the $YbGa_6$ peaks show only moderate overlap with that of the secondary phases. Thus, Rietveld refinements yielded structural parameters of reasonable accuracy even though there was no phase-pure sample available. There is no indication of incorporation of hydrogen into $YbGa_6$. We therefore consider the data given in Table 2 to describe the first refined crystal structure of $YbGa_6$.

Hydrogenation Behavior of $EuGa_2$. The hydrogenation of $EuGa_2$ was investigated with an autoclave experiment and with DSC experiments. The goal of the educt synthesis was only to produce a sample with $EuGa_2$ as a main phase, not to achieve pure single phase samples. Therefore, a sample consisting of 75.8(5) wt % $EuGa_2$ together with $EuGa_4$ (4.2(3) wt %), Eu_5Ga_9 (5.0(5) wt %), and Eu_3Ga_8 (15.0(3) wt %) was heated up to 300 °C under 90 bar of hydrogen gas pressure for 3 days in an autoclave. The resulting sample contained 85.1(8) wt % $EuGa_4$ and 14.9(8) wt % EuH_2 only.

The sample for the DSC experiments consisted of 63.6(5) wt % $EuGa_2$ together with Eu_5Ga_9 (5.6(4) wt %), $EuGa_4$ (17.2(3) wt %),

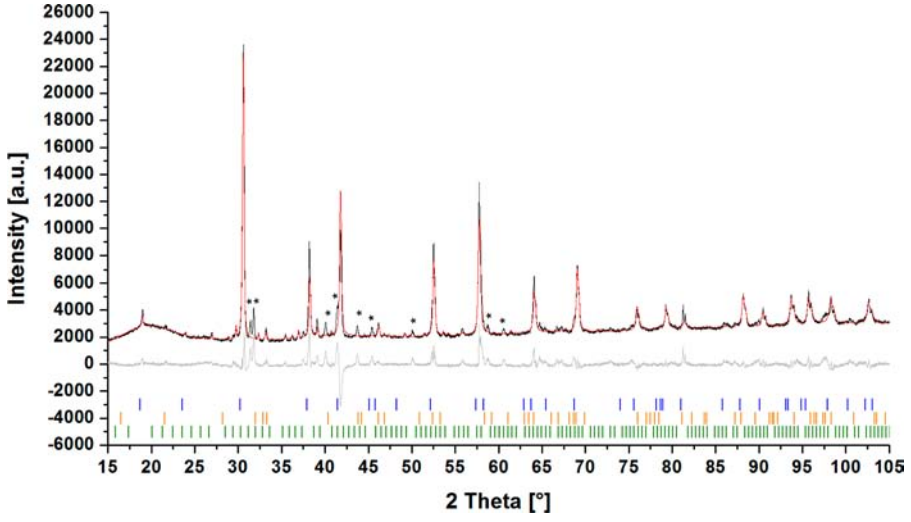

Figure 4. X-ray powder pattern (Cu Kα), with Rietveld refinement, of $SrGa_2$ before the hydrogenation. Bragg markers are, from top to bottom, $SrGa_2$, $SrGa_4$, and Sr_8Ga_7. Some of the peaks assigned to the unidentified phase are marked with an asterisk.

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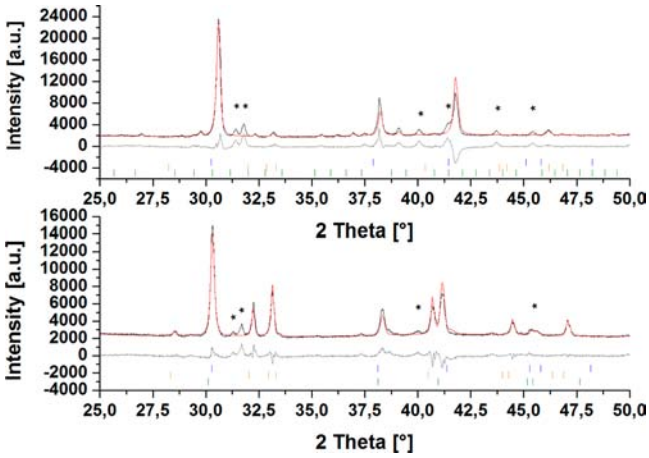

Figure 5. Detailed view of the X-ray powder pattern (Cu Kα) of $SrGa_2$ before (top panel) and after (bottom panel) hydrogenation in the DSC. Bragg markers are, from top to bottom, $SrGa_2$, $SrGa_4$, and Sr_8G_7 (top panel) and $SrGa_2H_2$ (bottom panel). Some of the peaks assigned to the unidentified phase are marked with an asterisk.

Eu_3Ga_8 (5.8(4) wt %) and EuO (7.9(2) wt %). Three conditions were chosen for the experiments: 50 bar of hydrogen gas pressure, 25 bar, and about 1 bar (i.e., purging only). Each experiment was heated to 180 °C and held for 2 h. Results of the experiments are given in Table 3.

Clearly, even without elevated hydrogen pressure, $EuGa_2$ decomposes into Ga-rich phases, so the weight fraction of these phases rises in all cases. Furthermore, EuO seems to be reduced by hydrogen at higher pressures. Thus, there is "missing" europium in all experiments; however, no further crystalline europium-containing phases are detected by XRD.

In conclusion, for EuGa2, results similar to those for $YbGa_2$ were obtained. $EuGa_2$ decomposes into $EuGa_4$ and EuH_2. The Zintl phases $EuGa_2$ and $YbGa_2$, in contrast to $SrGa_2$, show oxidative decomposition, which has already been observed in other compounds, for example, in Ca_3Ga_5, Li_3Al_2, and $SrZn_2$.[17]

Hydrogenation Behavior of $SrGa_2$. The hydrogenation of $SrGa_2$ was examined with the use of high pressure DSC. Conditions were 50 bar of H_2 pressure and a maximum temperature of 220 °C for 2 h. The heat flow curve is shown in Figure 3. The reaction starts at about 120 °C and has a strong increase of heat flow at about 180 °C. This is in agreement with the reaction conditions given by Björling et al. for the formation of $SrGa_2H_2$.[7] In the second cycle, no significant deviation from the baseline was detected, indicating that the hydrogenation reaction was already complete after the first cycle. The X-ray diffraction (Figure 4) of the $SrGa_2$ sample shows, before hydrogenation, weight fractions of $SrGa_2$ (90.3(4)%), $SrGa_4$ (1.5(2)%), and Sr_8Ga_7 (8.2(4)%), not including an unidentified phase.

After the hydrogenation, $SrGa_2$ was almost completely hydrogenated. The unidentified phase shows no reaction with hydrogen. Additionally, the weight fraction of $SrGa_4$ has increased, which may indicate the beginning of hydride decomposition. According to Björling et al.[7] $SrGa_2H_2$ decomposes at higher temperature into $SrGa_4$ and SrH_2. The binary hydride has perhaps not yet crystallized very well and therefore shows no peaks in the pattern. Weight fractions are 4.5(8)% for $SrGa_2$, 36.9(4)% for $SrGa_4$, and 58.6(6)% for $SrGa_2H_2$. The Sr_8Ga_7 phase was not found anymore. A representative part of the XRD pattern before and after the sample was hydrogenated is shown in Figure 5.

In addition to the hydrogenation in the DSC, an experiment with the same parameters was done in an autoclave. The result was similar to that of the hydrogenation in the DSC, and a Rietveld refinement (Figure 6) led to the following weight fractions: $SrGa_2$ 9.1(7), $SrGa_4$ 34.2(5), and $SrGa_2H_2$ 56.7(6).

In addition to the exploration and confirmation of the reaction conditions via DSC and autoclave, the hydride (deuteride) formation of $SrGa_2$ was investigated with in situ NPD. Deuterium gas was used to reduce the incoherent scattering of hydrogen. The reaction progress is shown in Figure 7. The sample was nearly phase pure, with little amounts of the aforementioned unidentified phase.

A comparison of the experimental with the calculated NPD patterns (Figure 7) shows that no intermediate product forms during the reaction, and the Zintl phase turns directly into the deuteride after half of the reaction time. At the end, $SrGa_4$ forms when the temperature reaches about 287 °C (small peaks rising at the end of the reaction time). In addition, the

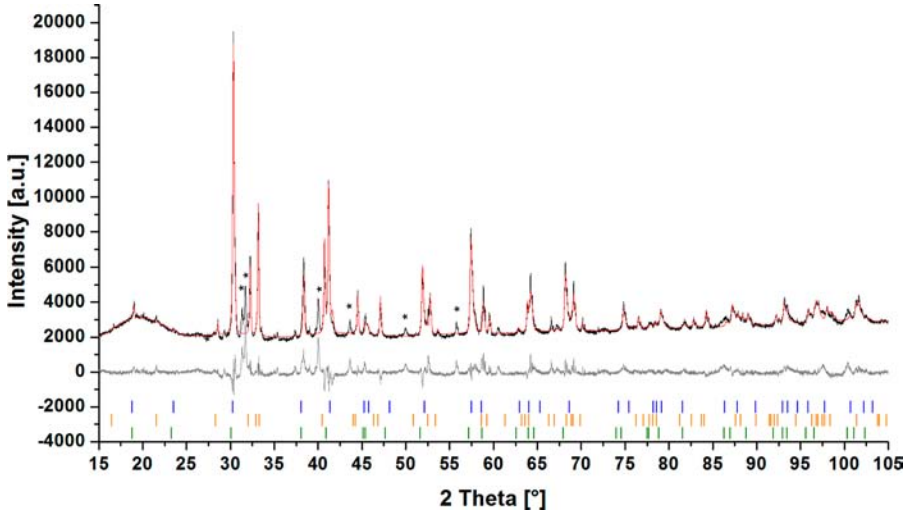

Figure 6. X-ray powder pattern (Cu Kα), with Rietveld refinement, of $SrGa_2$ after hydrogenation in the autoclave. Bragg markers are, from top to bottom, $SrGa_2$, $SrGa_4$, and $SrGa_2H_2$. Some of the peaks assigned to the unidentified phase are marked with an asterisk.

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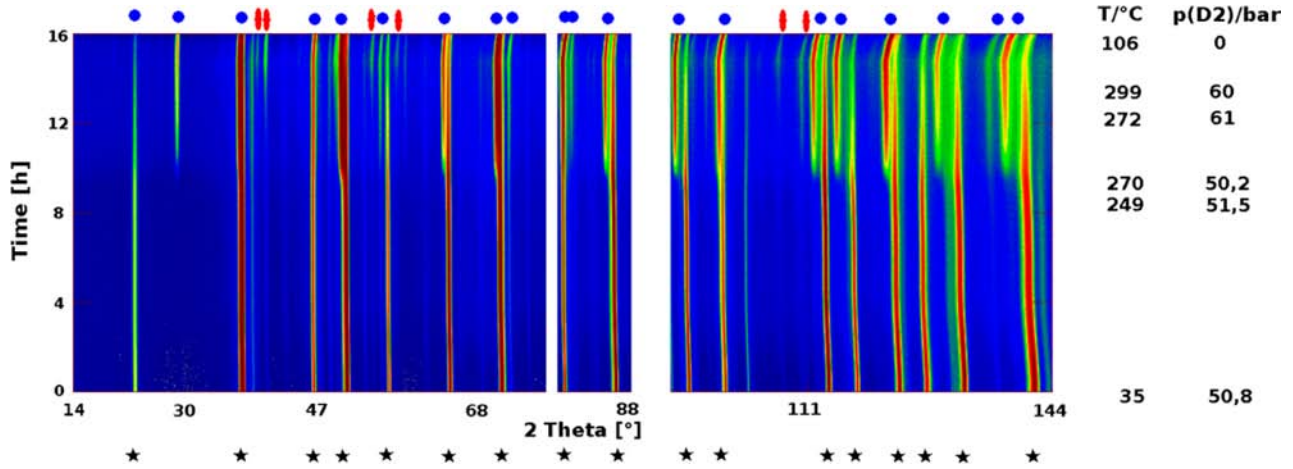

Figure 7. Time-dependent NPD patterns given in false colors (red = high intensity, blue = low intensity) showing the reaction progress of the deuteration of $SrGa_2$. At about 78° and 90° 2θ, peaks of the sapphire single crystal have been cut out. The refined wavelength is 1.86765(5) Å. The patterns were recorded with a time resolution of 2 min/scan. The black asterisks below the picture mark the position of the $SrGa_2$ peaks; above the picture, the peak positions of $SrGa_2D_2$ are marked by blue circles, and the $SrGa_4$ peak positions are marked by red ovals. On the righthand side, temperature and the pressure of deuterium are given.

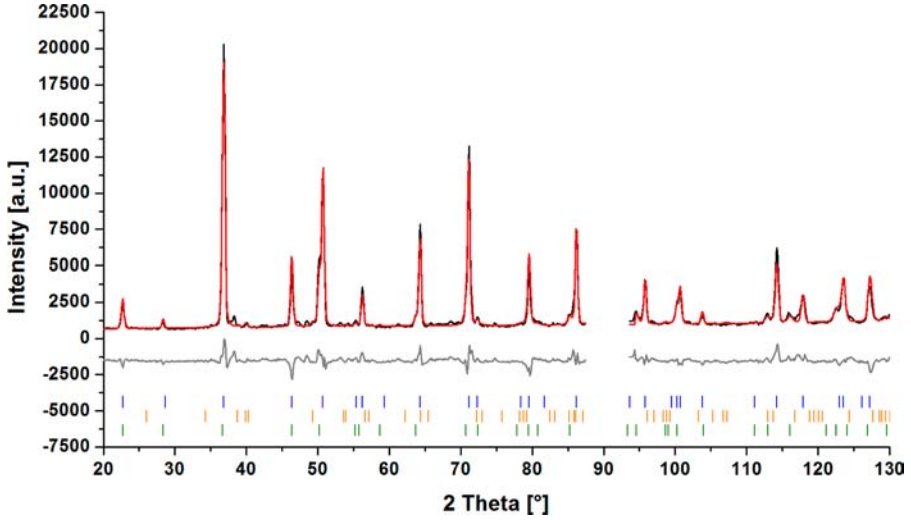

Figure 8. Neutron powder pattern (λ = 1.86765(5) Å) with Rietveld refinement at about 263 °C. Bragg markers are, from top to bottom, $SrGa_2$, $SrGa_4$, and $SrGa_2D_2$. The area cut out at about 90° in 2θ was excluded from the refinement and is the single crystal peak of the sapphire cell.

aforementioned unidentified phase continues to not react with hydrogen.

Figure 8 shows the Rietveld refinement at the start of the reaction, when the temperature is about 263 °C. The deuteride formation is strongly indicated by the rising peak at about 30° 2θ.

This is also confirmed by the nearly linear behavior of the lattice parameters, as shown in Figure 9. From the slope of the linear graphs in Figure 9, thermal expansion coefficients of 0.0029(1) $Å^3 \cdot °C^{-1}$ for the Zintl phase $SrGa_2$ and 0.0050(1) $Å^3 \cdot °C^{-1}$ for its deuteride can be determined, assuming, for each phase, a constant composition during the whole temperature range. These values represent typical thermal behavior for Zintl phases and hydrides (deuterides).[18] Hydrogen (deuterium) incorporation on the other hand has a much larger effect on unit cell volumes, as can be seen by a comparison of the respective values for $SrGa_2$ and $SrGa_2D_2$ (Figure 9 and Table 4). Therefore, we conclude that no deuterium uptake takes place before the formation of the deuteride begins. After cooling, the refined lattice parameters for the remaining $SrGa_2$ are not significantly different from those before the deuteration (a, 4.3483(2) → 4.3504(3) Å; c, 4.7404(2) → 4.7372(6) Å; both values at about 107 °C), which further supports this conclusion. Because of uncertainties in the temperature, temperature gradients in the sample, and the general underestimation of estimated standard uncertainties (e.s.u.) by the Rietveld method, we consider the differences of 4.2 times the standard uncertainty for the lattice parameter a and 4.0 for that of c to be not significant.

In Table 4 the structural parameters of the $SrGa_2D_2$ phase at three selected times during the reaction are given as an overview. In contrast to the findings of Björling et al.,[7] we found the deuterium occupation to be slightly below 1.0; however, the deviation from full occupation is only 3.3 e.s.u. In the same way, the positional parameters differ only slightly (maximum of 3.3 combined e.s.u. for z(D)), which indicates that the crystal structure is rather rigid and not very susceptible to deuterium

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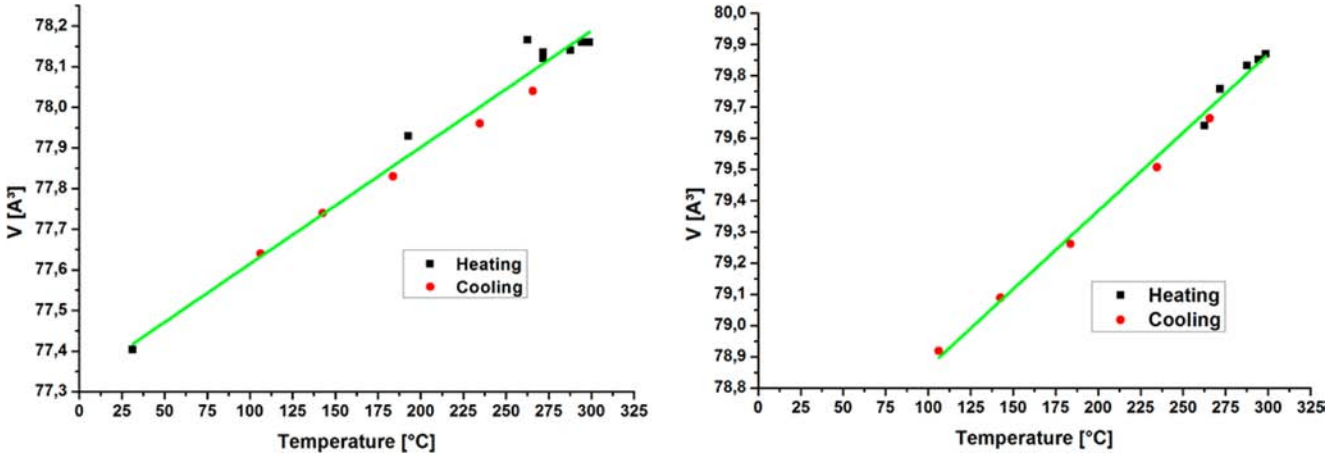

Figure 9. Refined unit cell volume of $SrGa_2$ (left) and $SrGa_2D_2$ (right) during the reaction progress. The data for heating (black) and cooling (red) are shown. Fitting each to a linear function yields V = 77.33(3) Å^3 + 0.0029(1) Å^3·°C^{-1}·T for $SrGa_2$ and V = 78.36(3) Å^3 + 0.0050(1) Å^3·°C^{-1}·T for $SrGa_2D_2$.

Table 4. Structural Parameters of the $SrGa_2D_2$ Phase at Selected Times from the Beginning of the Formation to the End of the Experiment

parameter	beginning of the reaction	end of heating	end of the experiment
T [°C]	272	299	106
p [bar]	54.5	60	1
space group		$P\bar{3}m1$ (No. 164)	
a [Å]	4.4077(2)	4.4098(2)	4.3969(2)
c [Å]	4.7409(4)	4.7429(3)	4.7138(2)
B (Sr), 1a: 0 0 0	2.8(4)	2.5(3)	1.8(2)
z (Ga), 2d: 1/3 2/3 z	0.477(2)	0.474(1)	0.472(1)
B (Ga)	1.2(2)	1.3(2)	0.7(2)
z (D), 2d: 1/3 2/3 z	0.125(2)	0.120(1)	0.115(1)
B (D)	2.1(3)	2.4(2)	1.6(2)
occupancy D-position	0.88(4)	0.90(3)	0.92(3)
d(Ga-D) [Å]	1.67(2)	1.680(9)	1.686(8)

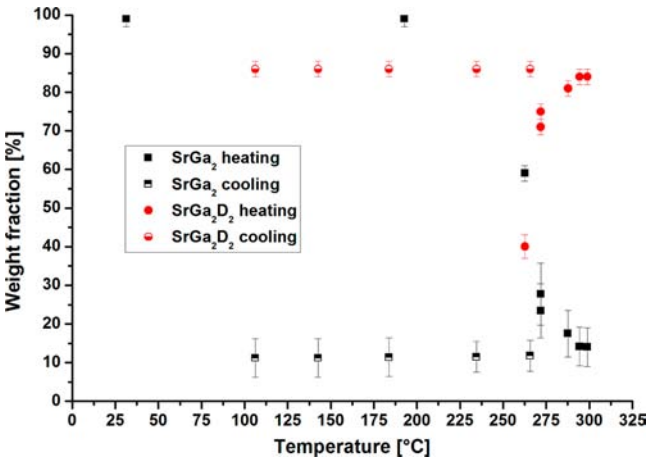

Figure 10. Weight fraction of $SrGa_2$ (black squares) and $SrGa_2D_2$ (red circles) during heating and cooling. The error bars represent 1 e.s.u.

gas pressure and temperature. Whether this hints of significant deuterium defects in the structure may be decided by independent experimental methods.

In Figure 10 the weight fractions of the gallide and the deuteride are shown; the fractions fall (and rise, respectively) rapidly after reaching a temperature of about 225 °C and remain nearly constant after the reaction. Clearly, the hydrogenation process is incomplete. We assume that the sample amount is too big for the laser beam and so the sample is not completely heated. Additionally, the thermal conductivity of the sample may be low enough to cause a temperature profile together with the falling temperature effect of the laser beam.

4. CONCLUSION

$SrGa_2$ is known to form the polyanionic hydride $SrGa_2H_2$, in which hydrogen is bound covalently to the gallium polyanion.[7] We have studied this hydrogenation reaction by in situ thermal analysis and in situ neutron diffraction. In the in situ DSC experiments at 50 bar hydrogen gas pressure, an exothermic peak with an onset around 125 °C indicates the formation of the polyanionic hydride. Temperature-dependent in situ NPD of the deuteration of $SrGa_2$ when heated to 300 °C in the pressure range of 50 ≤ $p(D_2)$ ≤ 61 bar shows a one-step reaction with no intermediate. $SrGa_2$ shows no sign of deuterium incorporation into its crystal structure before the reaction to $SrGa_2D_2$ takes place. The Rietveld refinement of the crystal structure of $SrGa_2D_2$ confirms the structural model reported earlier[7] for each temperature−pressure condition studied. The refined deuterium-occupation parameters do not vary significantly and are close to unity; that is, a stoichiometric or nearly stoichiometric composition is assumed for $SrGa_2D_2$. In contrast to $SrGa_2$, the analogous and structurally related gallides $EuGa_2$ and $YbGa_2$ do not form polyanionic hydrides in the investigated temperature−pressure ranges (20 °C ≤ T ≤ 220 °C, 5 bar ≤ $p(H_2)$ ≤ 54 bar). Hydrogenation of $YbGa_2$ leads to the formation of binary ytterbium hydride, $YbH_{2.67}$, $YbGa_4$, $YbGa_5$, and $YbGa_6$, depending on hydrogen gas pressure and temperature. The first refined crystal structural data for $YbGa_6$ in the tetragonal $PuGa_6$-type structure are presented. Hydrogenation of $EuGa_2$ leads to the formation of binary europium hydride, EuH_2, and $EuGa_4$. These are further examples of the oxidative decomposition of Zintl phases upon hydrogenation, which competes with the formation of Zintl phase hydrides or polyanionic hydrides.

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Notes

The authors declare no competing financial interest.

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