The crystal structure of Nb_3AuH_x

V.E. Antonov[a], E.L. Bokhenkov[a], B. Dorner[b], V.K. Fedotov[a], G. Grosse[c,*], A.I. Latynin[a],
F.E. Wagner[c], R. Wordel[c]

[a]*Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow district, Russia*
[b]*Institute Laue-Langevin, 15X, F-38042 Grenoble Cedex, France*
[c]*Physics Department E15, Technical University of Munich, D-85747 Garching, Germany*

Abstract

The A15 alloy Nb_3Au and its hydrides Nb_3AuH_x with $x=2.9$ and $x=4.2$ prepared at high hydrogen pressures were studied by neutron and X-ray diffraction. Their crystal structures were refined using the Rietveld method. Neutron diffraction showed that in both hydrides the hydrogen atoms occupy approximately 2.6 interstitial d sites of the 3 available per formula unit in the A15 metal lattice. The remaining hydrogen fills i sites. The X-ray examination revealed that the metal lattice of the Nb_3Au alloy and its hydrides was partly disordered, with $8\pm1\%$ of the regular Au positions randomly occupied by Nb. This atomic disorder is shown to be the cause of the observed maximum occupancy of d sites in the hydrides. © 1998 Elsevier Science S.A.

Keywords: Metal hydride; Neutron diffraction; X-ray diffraction

1. Introduction

The Nb_3Au alloy belongs to a large and well studied group of A_3B phases with the A15 structure, space group Pm3n. A number of these phases were shown to form hydrides, the maximum attained hydrogen contents being $Ti_3SbH_{2.1}$ [1], $Ti_3IrH_{3.9}$ [2], $Ti_3AuH_{2.8}$ [3], $V_3GaH_{1.9}$ [4], $Nb_3GeH_{0.86}$ and $Nb_3AlH_{2.18}$ [5], $Nb_3SnH_{2.2}$ [6], Nb_3RhH_6 [7], $Nb_3OsH_{4.1}$, $Nb_3IrH_{4.7}$, $Nb_3PtH_{5.1}$ and $Nb_3AuH_{4.3}$ [8]. All these A_3BH_x hydrides retain the A15 metal lattice, which exhibits the usual expansion with increasing hydrogen concentration.

One of the A15 hydrides, Nb_3SnH_x with $x\approx1$, was studied by neutron diffraction [9]. The hydrogen atoms were found to randomly occupy the 6-fold d positions of the space group Pm3n. Complete occupancy of these positions corresponds to a maximum hydrogen content of $x=3$. The maximum hydrogen solubility in the A15 alloys of Nb with the transition metals Rh, Os, Ir, Pt and Au, however, exceeds $x=4$, which implies that other interstitial sites are occupied either alone or in addition to the d sites.

Of this family, the Nb_3AuH_x hydrides were chosen for the present structure study because of the low lower limit of their homogeneity range of compositions [8]. This made it possible to prepare hydrides with hydrogen contents both

below ($x\approx2.9$) and substantially above ($x\approx4.2$) the value of $x=3$ characteristic of the complete occupancy of the d sites. Moreover, a [197]Au Mössbauer investigation of Nb_3AuH_x [6] has revealed an interesting phenomenon: the spectrum of the Nb_3Au starting alloy was found to consist of an absorption line with a pronounced shoulder, which was no longer observed in the spectra of the $Nb_3AuH_{2.8}$ and $Nb_3AuH_{4.2}$ hydrides. As the spectrum of the Nb_3Au alloy should consist only of the single absorption line arising from Au on c positions with m3 symmetry in the ideal A15 structure, a careful investigation of the structure of this alloy and of the metal lattice of its hydrides seemed desirable.

The hydrogen positions in the crystal structure of the Nb_3AuH_x hydrides were determined by neutron diffraction, whereas the investigation of the Nb and Au distribution over the sites of the A15 metal lattice required a separate X-ray examination because of the small difference between the neutron scattering lengths of niobium (7.054 Å) and gold (7.90 Å) [10].

2. Sample preparation and experimental details

As in Refs. [6] and [8], the starting material was a $Nb_{2.96}Au_{1.04}$ alloy (Nb_3Au for brevity hereafter) melted from the elements of 99.99 wt % purity in a levitation

*Corresponding author.

induction furnace under an argon atmosphere and then annealed in a vacuum of 10^{-4} Pa at 1100 °C for 24 h and cooled together with the furnace. The hydrides, $Nb_3AuH_{2.88\pm0.06}$ and $Nb_3AuH_{4.20\pm0.08}$, were prepared by exposing 0.3 mm thick plates, cut from the ingot with an abrasive wire saw, to hydrogen pressures of 0.85 and 5.1 GPa, respectively, at 325 °C for 24 h with subsequent cooling to 100 K in the high pressure cell. The method of hydrogenation is described in more detail elsewhere [11].

At ambient pressure, the sample with $x = 4.20$ began to lose hydrogen on warming to about -20 °C. At room temperature, the hydrogen content of this sample decreased to $x \approx 2.8$ in a few hours; no release of hydrogen from the sample with $x = 2.88$ was observed at room temperature in the course of a week. The hydrogen content was determined by hot extraction into a calibrated volume at temperatures up to 500 °C using about 5 mg of each sample.

When not in use, the hydrides were stored in liquid nitrogen. Prior to the diffraction measurements, they were ground in an agate mortar under liquid nitrogen in order to avoid texture effects. The starting Nb_3Au alloy was ground at room temperature.

The neutron diffraction experiments were performed at 120 K, using the D20 instrument at the ILL, Grenoble, with neutrons of a wavelength of $\lambda = 1.295$. The powder diffraction patterns were scanned in steps of 0.1° in 2Θ. Both fixed-time and monitor counting schemes were employed. About 1 g samples of Nb_3Au, $Nb_3AuH_{2.88}$ and $Nb_3AuH_{4.20}$ were used.

The X-ray diffraction experiments were performed at room temperature for the Nb_3Au and $Nb_3AuH_{2.88}$ samples using a SIEMENS D-500 diffractometer and monochromated CuK_{α_1} radiation. The patterns were scanned in steps of 0.02° in 2Θ. The $Nb_3AuH_{4.20}$ sample was not measured by X-ray diffraction because it would have lost hydrogen at ambient temperature.

The neutron and X-ray data were analysed using the Rietveld profile refinement technique implemented in the DBWS-9411 computer program [12], which allows the simultaneous refinement of several phases.

3. Results

3.1. X-ray diffraction

The analysis of the X-ray diffraction spectrum of the initial Nb_3Au alloy (Fig. 1) showed that, in addition to the A15 phase, the sample contained impurities which were identified as the tetragonal Nb_3Au_2 and cubic β-Mn type $Nb_{11}Au_9$ compounds described in [13]. The relative amount of the impurities given by the Rietveld refinement agree with the notion [14] that the maximum gold content of the A15 phase corresponds to the composition
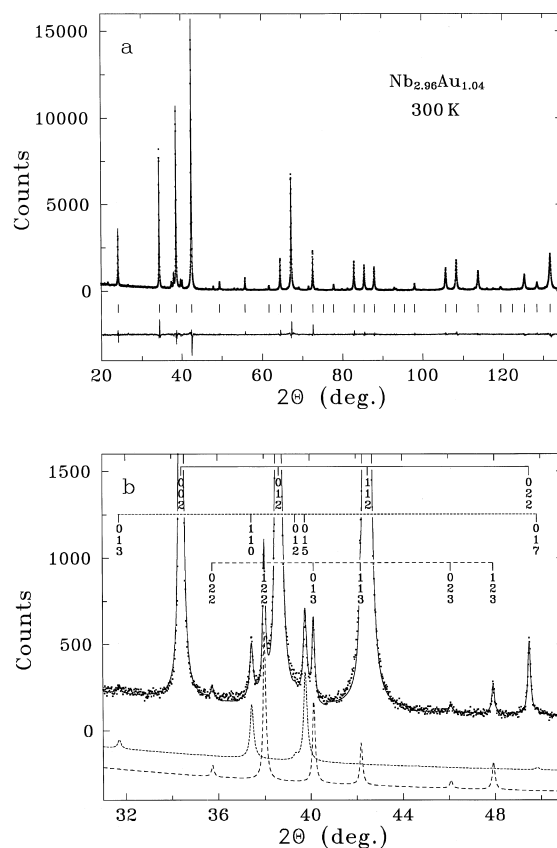

Fig. 1. X-ray diffraction pattern of the $Nb_{2.96}Au_{1.04}$ alloy at 300 K and results of its Rietveld refinement involving the $Nb_{2.98}Au_{1.02}$, Nb_3Au_2 and $Nb_{11}Au_9$ phases (Table 1). The experimental data are shown by dots, the calculated profile as a solid line. The difference curve is presented in the lower part of Fig. 1(a), the contributions from the Nb_3Au_2 (short-dashed curve) and $Nb_{11}Au_9$ (long-dashed curve) in the lower part of Fig. 1(b).

$Nb_{2.98}Au_{1.02}$. The studied sample can thus be considered as

$$Nb_{2.96}Au_{1.04} \approx 0.97Nb_{2.98}Au_{1.02} + 0.02Nb_3Au_2 + 0.002Nb_{11}Au_9.$$

The refined parameters for the component and the impurity phases are listed in Table 1. Since variations of the site occupancy numbers, ω, and the thermal factors of the atoms, B, influence the calculated spectrum in much the same manner, and since their values are therefore strongly correlated, we used the B values determined from the neutron diffraction data (Table 2), which are insensitive to the ω values because of the similar neutron scattering lengths of Nb and Au [10].

As Table 1 shows, the A15 structure of the $Nb_{2.98}Au_{1.02}$ phase exhibits a substantial degree of chemical disorder: about 8% of the 2-fold a sites normally occupied by gold are randomly occupied by Nb and about 3% of the 6-fold c niobium sites are occupied by Au.

The X-ray diffraction pattern of $Nb_3AuH_{2.88}$ is similar to that of Nb_3Au. Its Rietveld refinement converged to

Table 1

Positional parameters (x, y, z) and thermal factors (B) for the constituent phases of the $Nb_{2.96}Au_{1.04}$ alloy according to the Rietveld profile refinement analysis of the X-ray diffraction data collected at 290 K. N is the number of atoms per formula unit, ω is the site occupancy, M is the number of formula units per unit cell, R_p and R_{ex} are the obtained and expected profile factors.

Phase	Atom	Site	x	y	z	B (Å^2)	ω	N
$Nb_{2.98}Au_{1.02}$	Au	2(a)	0.000	0.000	0.000	0.32	0.92	0.92
Pm3n, $M=2$	Nb	6(c)	0.250	0.000	0.500	0.42	0.97	2.90
$a=5.201$ Å	Nb	2(a)	0.000	0.000	0.000	0.42	0.08	0.08
	Au	6(c)	0.250	0.000	0.500	0.32	0.03	0.10
Nb_3Au_2	Nb	2(a)	0.000	0.000	0.000	0.42	1.00	1.00
I4/mmm, $M=2$	Nb	4(e)	0.000	0.000	0.400	0.42	1.00	2.00
$a=3.391$ Å, $c=15.18$ Å	Au	4(e)	0.000	0.000	0.200	0.32	1.00	2.00
$Nb_{11}Au_9$	Au	8(c)	0.061	0.061	0.061	0.32	1.00	8.00
P4$_1$32, $M=1$	Nb	12(d)	0.125	0.206	0.456	0.42	0.92	11.00
$a=7.092$ Å	Au	12(d)	0.125	0.206	0.456	0.32	0.08	1.00

$R_p/R_{ex}=7.6/9.9$.

$R_p/R_{ex}=7.0/6.0$. The impurity fractions and the occupation numbers of the a and c sites in the A15 phase did not change upon hydrogenation. The lattice parameter of the $Nb_{11}Au_9$ phase did not change either, whereas the lattice parameters of the $Nb_{2.98}Au_{1.02}$ and Nb_3Au_2 phases increased from the values given in Table 1 for the non-hydrogenated material to $a=5.415$ Å and to $a=3.452$ Å, $c=15.30$ Å, respectively.

The impurities in the samples were found to practically not affect the accuracy of the Rietveld fit for the A15 phase. Their investigation allows conclusions about the interaction of hydrogen with Nb_3Au_2 and $Nb_{11}Au_9$ which have not yet been studied. The dissolution of hydrogen in metals and alloys with a variety of crystal structures, including the A15 structure [8] of Nb_3Au, has been found to cause the lattice to expand by about 2–3 Å^3 per

dissolved hydrogen atom [11,15]. Within the experimental error, the lattice parameter of the $Nb_{11}Au_9$ phase did not change upon hydrogenation of the sample. This indicates that its hydrogen content did not exceed few atomic per cent. The lattice expansion of the Nb_3Au_2 phase in the $Nb_3AuH_{2.88}$ sample corresponds to the formation of a hydride with the estimated composition $Nb_3Au_2H_{\approx1.5}$.

From the equation

$$Nb_{2.96}Au_{1.04}H_{2.88} \approx 0.97Nb_{2.98}H_x + 0.02Nb_3Au_2H_{1.5}$$
$$+ 0.002Nb_{11}Au_9$$

it follows that $x\approx2.94$. The hydrogen content of the A15 phase, $x\approx2.9$, thus did not differ from the average hydrogen content of the sample, $\bar{x}=2.88\pm0.06$, within the experimental accuracy.

Table 2

Positional parameters (x, y, z) and thermal factors (B) for the $Nb_{2.98}Au_{1.02}$ compound and its hydrides according to the Rietveld profile refinement analysis of the neutron diffraction data. N is the number of atoms per formula unit, ω is the site occupancy, R_p and R_{ex} are the obtained and expected profile factors. Cubic structure, space group Pm3n, $T=120$ K.

| Phase	Atom	Site	x	y	z	B (Å^2)	ω	N
$Nb_{2.98}Au_{1.02}$	Au	2(a)	0.000	0.000	0.000	0.32	0.92	0.92
$a=5.192$ Å	Nb	6(c)	0.250	0.000	0.500	0.42	0.97	2.90
$R_p/R_{ex}=2.9/4.2$	Nb	2(a)	0.000	0.000	0.000	0.42	0.08	0.08
	Au	6(c)	0.250	0.000	0.500	0.32	0.03	0.10
$Nb_{2.98}Au_{1.02}H_{2.9}$	Au	2(a)	0.000	0.000	0.000	0.33	0.92	0.92
$a=5.409$ Å	Nb	6(c)	0.250	0.000	0.500	0.42	0.97	2.90
$R_p/R_{ex}=2.9/4.1$	Nb	2(a)	0.000	0.000	0.000	0.42	0.08	0.08
	Au	6(c)	0.250	0.000	0.500	0.33	0.03	0.10
	H$_d$	6(d)	0.250	0.500	0.000	1.03	0.85	2.56
	H$_i$	16(i)	0.217	0.217	0.217	1.03	0.02	0.15
$Nb_{2.98}Au_{1.02}H_{4.2}$	Au	2(a)	0.000	0.000	0.000	0.39	0.92	0.92
$a=5.465$ Å	Nb	6(c)	0.250	0.000	0.500	0.48	0.97	2.90
$R_p/R_{ex}=5.4/4.1$	Nb	2(a)	0.000	0.000	0.000	0.48	0.08	0.08
	Au	6(c)	0.250	0.000	0.500	0.39	0.03	0.10
	H$_d$	6(d)	0.250	0.500	0.000	1.28	0.86	2.58
	H$_i$	16(i)	0.215	0.215	0.215	1.85	0.18	1.42
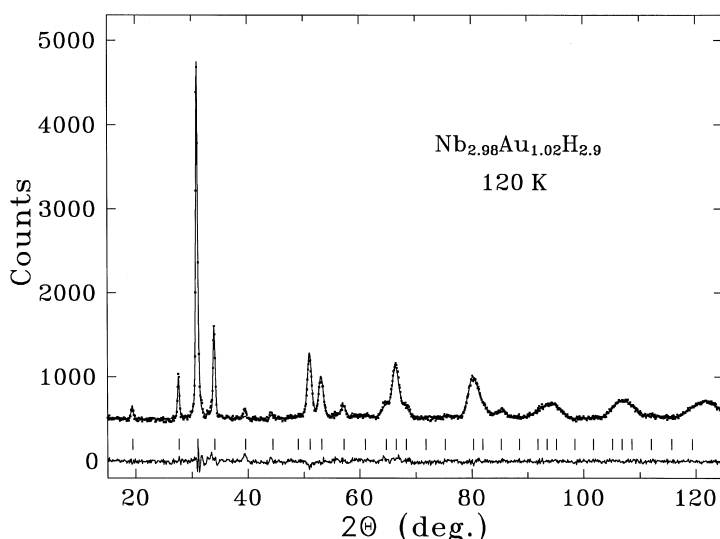
Fig. 2. The neutron diffraction pattern of the $Nb_{2.96}Au_{1.04}H_{2.88}$ sample at 120 K. The background and contributions from $Nb_3Au_2H_{1.5}$ and $Nb_{11}Au_9$ resulting from the Rietveld refinement have been subtracted. The experimental data are shown by dots, the fitted and difference curves as solid lines.

3.2. Neutron diffraction

The neutron diffraction patterns of both studied $Nb_{2.96}Au_{1.04}H_x$ hydrides (Fig. 2 presents an example) contained no new lines compared to that of the initial $Nb_{2.98}Au_{1.02}$ phase. The crystal structures of the hydrides therefore belong to the same Pm3n space group as the structure of the initial A15 compound. Applying the Rietveld profile refinement method to the neutron data we found the stable solutions listed in Table 2. Hydrogen occupies d and i positions in the A15 metal lattice of both hydrides, with nearly the same occupancy of the d positions. The calculated total hydrogen content of the hydrides, $N_d + N_i$, is in a satisfactory agreement with the data of hot extraction. The sites occupied by different atoms in the resultant hydride structure are shown in Fig. 3, the nearest interatomic distances are given in Table 3.

The results of the Rietveld profile analysis (Table 2) are in agreement with qualitative estimates from the relative line intensities, I_{hkl}, in the neutron diffraction spectra of $Nb_{2.98}Au_{1.02}$ and its two hydrides. For example, as is seen

Table 3
Nearest interatomic distances (in Å) for Nb_3AuH_x hydrides with the a and x_i values from Table 2.

Atom-Atom	Formula	$Nb_3AuH_{2.9}$	$Nb_3AuH_{4.2}$
Au-Nb, Au-H_d	$a\sqrt{5}/4$	3.024	3.055
Au-H_i	$a\sqrt{3}x_i$	2.033	2.035
Nb-H_d	$a\sqrt{2}/4$	1.912	1.932
Nb-H_i, H_d-H_i	$a\sqrt{3(1/4 - x_i)^2 + 1/8}$	1.937	1.960

from the formulae in Fig. 4, I_{012} should increase whereas I_{002} and I_{112} should decrease with increasing occupancy of the d sites. Increasing occupancy of the i sites leads to an increase in I_{002}, but does not affect I_{012} and I_{112}. The experiment shows that the formation of $Nb_{2.98}Au_{1.02}H_{2.9}$ is accompanied by an increase in I_{012} and a decrease in I_{112}, which is possible only if the d positions are filled. Both I_{012} and I_{112} are virtually the same for $Nb_{2.98}Au_{1.02}H_{2.9}$ and $Nb_{2.98}Au_{1.02}H_{4.2}$, indicating the same occupancy of the d sites. I_{002}, however, is higher for $Nb_{2.98}Au_{1.02}H_{4.2}$, which can be attributed to a higher occupancy of the i sites.
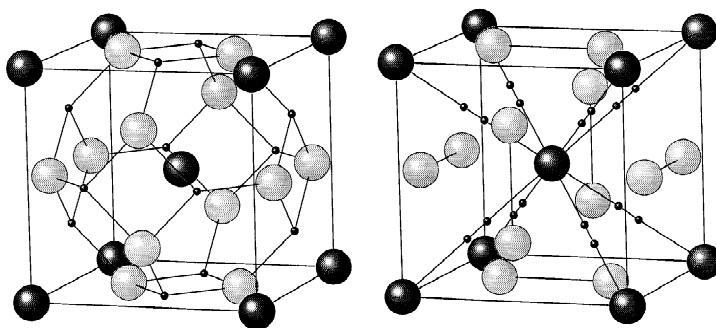

Fig. 3. Crystal structure of Nb_3AuH_x. Au atoms are shown as dark spheres, Nb atoms as light spheres and the H interstices as small black spheres. In the left and right unit cell the interstitial d and i positions, respectively, are shown.
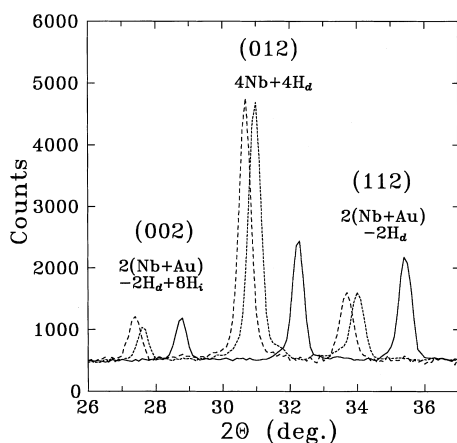
Fig. 4. A portion of the neutron diffraction spectra of the $Nb_{2.96}Au_{1.04}$ (solid line), $Nb_{2.96}Au_{1.04}H_{2.9}$ (short-dashed line) and $Nb_{2.96}Au_{1.04}H_{4.2}$ (long-dashed line) phases at 120 K with subtracted background. The spectra are normalised to the number of formula units and to the intensity of the incident neutron beam. The formulae under the line indices give approximate contributions from different atoms to the corresponding structure amplitudes.

The lattice parameter of the $Nb_{11}Au_9$ impurity is the same in the initial and in the two hydrided samples. This suggests a low hydrogen solubility in $Nb_{11}Au_9$ at 325 °C and pressures up to 5.1 GPa. It is highly improbable that the samples prepared under these conditions could have lost hydrogen before the neutron diffraction experiments, because they were quenched to 100 K in the high pressure cell and never heated above 120 K. No other hydrides studied so far lost hydrogen at temperatures that low [11].

The neutron diffraction values of the lattice parameters of the $Nb_3Au_2H_x$ impurity in the $Nb_3AuH_{2.88}$ sample agree with those determined from the X-ray data, if they are corrected for thermal expansion due to the different temperatures of measurement (120 and 300 K). This confirms that Nb_3Au_2 forms a hydride of the approximate composition $Nb_3Au_2H_{1.5}$ at 325 °C and a hydrogen pressure of 0.85 GPa, and that this hydride is thermally stable at ambient pressure and room temperature.

When calculating the contribution from the $Nb_3Au_2H_{1.5}$ impurity to the neutron diffraction spectrum of the $Nb_3AuH_{2.88}$ sample, we assumed hydrogen to occupy the 8-fold g positions of the space group I4/mmm. The g positions are tetrahedral interstitial sites with only Nb atoms as the nearest neighbours. The fraction of the $Nb_3Au_2H_{1.5}$ phase in the sample was, however, too small for a reliable test whether this assumption is correct.

No lines attributable to a $Nb_3Au_2H_x$ impurity were observed in the diffraction pattern of the $Nb_3AuH_{4.20}$ sample. Most probably, the Nb_3Au_2 phase in this sample formed a higher hydride with a different crystal structure, whose strongest diffraction lines are hidden under the peaks of the main $Nb_{2.98}Au_{1.02}H_{\approx4.2}$ phase. The presence of such a phase can also explain the fact that for the $Nb_3AuH_{4.20}$ sample the R_p/R_{ex} ratio of the Rietveld fit,

which involved only two phases, $Nb_{2.98}Au_{1.02}H_x$ and $Nb_{11}Au_9$, turned out to be worse than for the two other samples (Table 2).

4. Discussion

The arrangement of hydrogen atoms in the crystal structure of the $Nb_{2.98}Au_{1.02}H_{2.9}$ and $Nb_{2.98}Au_{1.02}H_{4.2}$ hydrides suggested by the Rietveld refinement of their neutron diffraction patterns (Table 2 and Fig. 3) appears sound on both chemical and structural grounds. As to the chemical grounds, one should take into account the affinity of hydrogen to niobium, the repulsive interaction of hydrogen with Au atoms dissolved in transition metals [16] and the blocking effect requiring that the distance between hydrogen atoms in a metal should not be much less than 2 Å [17]. The structural restrictions are that hydrogen should occupy only interstitial sites allowing accommodation of atoms with the effective radius of 0.56–0.60 Å [17] and that the hydrogen positions should conform to the Pm3n space group.

As is seen from Table 2 and Fig. 3, in the Nb_3AuH_x hydrides the gold atoms at the a positions form a bcc lattice with $a \approx 5.4$ Å. The niobium atoms occupy half of the tetrahedral interstitial sites (c positions) in this bcc lattice. The other half of the tetrahedral sites (d positions) lie at the centres of tetrahedrons formed by nearest-neighbour Nb atoms at a distance of about 1.91–1.93 Å, which makes the d sites similar to the tetrahedral positions occupied by hydrogen in niobium hydride [18]. The nearest Au atoms being about 3 Å away, the d positions therefore look most favourable for hydrogen occupancy. This agrees with the results of the Rietveld analysis showing that d sites in the $Nb_{2.98}Au_{1.02}H_x$ hydrides are the first to be filled with hydrogen.

The distance between the neighbouring d sites in the $Nb_{2.98}Au_{1.02}H_x$ hydrides is more than 2.7 Å, therefore no blocking effects are expected. The complete filling of these sites would correspond to a hydride composition of $Nb_{2.98}Au_{1.02}H_3$. The experiments, however, give a value of $N_d = 2.56$–2.58 for both studied hydrides (Table 2). The observed maximum occupancy of the d sites can be explained by the atomic disorder in the metal sublattice of the hydrides if one assumes that d sites with one or more Au atoms replacing Nb on a nearest-neighbour c site cannot be occupied by hydrogen atoms. Assuming a random occupation of c sites by Au atoms, the number of such Au neighbours of a d site is binomially distributed. The number of d sites that can be occupied by hydrogen atoms is thus

$$N_d^{calc} = 3 \cdot \binom{4}{0}(1-c)^4 = 2.62 \pm 0.03,$$

where $c = 0.033 \pm 0.003$ is the concentration of Au atoms on c sites as derived from the X-ray study. The calculated

limiting value, $N_d^{\text{calc}} = 2.62$, is in excellent agreement with the N_d values resulting from the Rietveld fit of the neutron diffraction data (Table 2).

The incomplete filling of the d sites also agrees with the presence of a wide plateau with $x \approx 2.8$–2.9 in the pressure dependence of the hydrogen solubility in Nb_3Au at 325 °C [8]. This plateau extends from about 0.2 to 1.5 GPa and indicates a change in the type of the interstitial sites predominantly occupied by the hydrogen added at higher pressures.

According to the Rietveld analysis (Table 2), these sites are the 16-fold tetrahedral i sites with the positional parameter $x_i \approx 0.22$. These sites lie in pairs along the <111> space diagonals (Fig. 3) inside asymmetrical tetrahedra of three Nb atoms at a distance of 1.94–1.96 Å and a Au atom at 2.03–2.04 Å (Table 3). The distance between the two i positions within a pair is $a\sqrt{3}\,(1/2 - 2x_i) \approx 0.62$–0.66 Å, which is much less than 2 Å. Therefore, the two i sites of a pair cannot be occupied by hydrogen at the same time due to the blocking effect [17]. Occupancy of half of the i sites corresponds to $N_i^{\max} = 4$.

The maximum value of $N_i \approx 1.4$ obtained experimentally in $Nb_{2.98}Au_{1.02}H_{4.2}$ (Table 2) is much smaller. One therefore has to assume that an additional blocking mechanism prevents that more i sites become occupied. A conceivable possibility is that the occupation of i sites causes a displacement of the neighbouring Au atom, which prohibits the occupation of other neighbouring i sites by hydrogen.

If the local distortions caused by hydrogen atoms occupying asymmetric i sites are random in character, the cubic symmetry of the mean lattice as observed by diffraction methods remains unchanged, but random static displacements of the atoms contribute to the Debye–Waller factor. This idea is therefore confirmed by the nearly equal B values of Au and Nb in $Nb_{2.98}Au_{1.02}H_{2.9}$ and $Nb_{2.98}Au_{1.02}$ and the considerable increase in the B values of all atoms in $Nb_{2.98}Au_{1.02}H_{4.2}$ (Table 2).

There are still other interstitial sites in the A15 metal lattice, namely the 24-fold k positions situated inside asymmetric tetrahedra formed by three Nb atoms and one Au atom. The k positions are occupied by hydrogen in β-UH_3, which has the A15 metal lattice with uranium on both a and c sites [19]. The volume of k sites in Nb_3AuH_x is, however, approximately two times smaller than in UH_3 and these sites are therefore unlikely candidates for hydrogen uptake.

5. Conclusions

Within the homogeneity range of the Nb_3AuH_x hydrides, i.e., from $x \approx 2.8$ to $x \approx 4.3$, hydrogen occupies two types of interstitial positions, d and i, in the A15 metal lattice. The occupancy of the d positions is nearly independent of the total hydrogen content and equal to

approximately 85%, which corresponds to $N_d \approx 2.6$ hydrogen atoms per formula unit. The occupancy of the i sites increases from about 2% for the hydride with $x = 2.9$ to about 18% for that with $x = 4.2$, the latter corresponding to $N_i \approx 1.4$.

The A15 metal lattice of the studied Nb_3Au compound and its hydrides was partly disordered. About 8% of the regular Au positions were occupied by Nb and about 3% of the regular Nb positions by Au. This atomic disorder explains the observed maximum concentration, $N_d \approx 2.6$, of hydrogen on d sites under the assumption that hydrogen does not occupy d sites with one or more nearest-neighbour Nb atoms replaced by Au atoms.

The atomic disorder can also account for the complex ^{197}Au Mössbauer spectrum of the initial Nb_3Au sample reported earlier [6]. Gold atoms occupying c sites expectedly have different hyperfine parameters than those on regular a sites. Furthermore, even Au atoms on regular a sites have different environments arising from the replacement of Nb with Au atoms on neighbouring c sites. The Mössbauer spectrum can therefore be expected to be complex and can be interpreted correctly only by taking the atomic disorder into account. The results of its analysis will be published later [20] in the framework of a more general Mössbauer investigation of the Nb-Au-H system.

An investigation of impurities in the Nb_3AuH_x samples showed that the hydrogen solubility in the $Nb_{11}Au_9$ compound does not exceed few atomic per cent at 325 °C and pressures up to 5.1 GPa, while the Nb_3Au_2 compound forms a hydride with an approximate composition of $Nb_3Au_2H_{1.5}$ at 325 °C and 0.85 GPa. This hydride is thermally stable at ambient conditions.

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