PHYSICAL REVIEW B **66**, 144423 (2002)

Effect of hydrogenation on crystal structure and magnetic properties of UTSi (T=Pd,Ni) intermetallics

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(Received 21 March 2002; published 31 October 2002)

The crystal structure and magnetic properties of UPdSi and UNiSi hydrides synthesized at $T=923$ K and hydrogen pressure $p=130$ bars were studied. Hydrogenation results in modification of the lattice symmetry from the orthorhombic (TiNiSi structure type) to hexagonal (ZrBeSi type) and in the increase of magnetic ordering temperatures in both compounds. UPdSiH$_x$ orders antiferromagnetically at $T_N=46$ K compared to $T_N=31$ K for UPdSi. UNiSiH$_x$ is a ferromagnet with $T_C=98$ K, whereas UNiSi is an antiferromagnet with $T_N=85$ K. The observed changes are attributed to the increased uranium-uranium spacing in the hydrides.

DOI: 10.1103/PhysRevB.66.144423 PACS number(s): 75.30.−m, 61.10.−i, 81.40.Rs

I. INTRODUCTION

Hydrogenation of uranium-based intermetallic compounds, leading to enlarged U-U spacing and modified bonding conditions, is an interesting probe of 5f-electron systems. Magnetic properties, which are very sensitive to the degree of the 5f localization, are important indicators of electronic structure variations.[1] In addition, hydride studies are also relevant for hydrogen storage issues.[2]

However, not all U intermetallics absorb hydrogen under the usual laboratory conditions. Attempts to hydrogenate the UTX materials (T=transition metal atom, $X=p$-electron element) crystallizing in the ZrNiAl structure type were successful in a few cases only,[3,4] although rare-earth (R) isotypes absorb H$_2$ willingly.[5]

In this work we describe the synthesis and study of two hydrides based on UPdSi and UNiSi, representatives of the UTX compounds crystallizing in the orthorhombic TiNiSi structure type.

UPdSi was thoroughly investigated by several groups by means of x-ray and neutron diffraction, magnetization, and transport measurements.[6–11] It was established that UPdSi crystallizes in the TiNiSi-type of structure (space group $Pnma$) having the shortest inter-uranium spacing $d_{U\text{-}U}\approx 350$ pm. This value is close to the critical region between localized and itinerant 5f-electron behavior.[1] UPdSi is an antiferromagnet, which undergoes two magnetic phase transitions at $T_N=29$ K and $T_1=10$ K according to Ref. 6 or $T_N=33$ K and $T_1=27$ K according to Refs. 8 and 9. It has also two metamagnetic transitions at $\mu_0H_{c_{1,2}}=4$ T and 7 T ($T=4.2$ K), respectively, at which the antiferromagnetic coupling of the U moments is gradually broken.[9]

The other intermetallic compound studied, UNiSi, has also the TiNiSi-type of unit cell, but with a lower $d_{U\text{-}U}\approx 330$ pm.[12] It shows more complicated magnetic behavior and three magnetic phase transitions are reported for this compound. Magnetization measurements point to antiferromagnetic ordering at $T_N=80$ K followed by another phase transition at $T_1=7.5$ K.[6] Specific heat studies revealed an additional transition at $T=18$ K.[8] The nonzero spontaneous magnetization observed at lower temperatures is associated with ferromagnetic ordering or the presence of a ferromagnetic component. Below $T=10$ K, the magnetization curves have a complicated S-shaped character, which indicates that in the ground state UNiSi is a ferrimagnet or a ferromagnet with a canted structure.[10] The low value of the effective moment $\mu_{eff}=2.1$ μ_B/f.u. given in Ref. 6, as well as the poor approach to saturation of the "free-powder" magnetization curve even in the field of 35 T ($M_{35T}=0.52$ μ_B/f.u.), points to strongly delocalized U 5f states (see Ref. 13 and references therein). The ferromagnetic component disappears above $T=18$ K.[10] The negative value of the paramagnetic Curie temperature $\Theta_P=-13$ K estimated from the magnetic susceptibility measured above 80 K points to antiferromagnetic interactions dominating in the system.[6]

II. EXPERIMENTAL DETAILS

The parent compounds were obtained by arc melting of stoichiometric amounts of constituents under Ar atmosphere. The phase composition was checked by means of x-ray diffraction on the Siemens D-500 diffractometer equipped with Cu (used for UPdSi-H) and Co (used for UNiSi-H) anodes. Prior to exposing the samples to hydrogen, they were crushed to submillimeter particles and their surface was activated by annealing for 2 h at $T=473$ K in the dynamic vacuum of $p=2\times10^{-5}$ mbar. Hydrogenation was performed at $T=923$ K under hydrogen pressure $p_{H_2}=130$ bars. Several attempts to obtain the hydrides at lower H$_2$ pressure and lower temperature were unsuccessful. The absorption process was monitored by the pressure variations in closed calibrated volume. It was considered to be completed after the pressure drop finished. The crystal structures of the synthesized products were checked by x-ray diffraction. It was not possible to determine precisely enough the hydrogen content directly during the synthesis, so the weight change due to the H$_2$ uptake was used to estimate the hydride composition. It appeared that the samples of both compounds contain about 2 H atoms/f.u. immediately after the synthesis, and with time they tend to decompose losing about 1 H atoms/f.u. In order to decompose the hydrides completely, they were heated to 973 K in the closed evacuated volume. The pressure increase due to the H_2 release usually started around 723 K and reached the saturation until 923 K. To ensure that all hydrogen was removed from the sample it was heated further to 973 K, held at that temperature, and then the reactor was pumped down to <3 mbar. The sample was cooled at continuous pumping. The products of decomposition were again analyzed by x-ray diffraction.

Magnetic measurements on fine powders with particles fixed in a random orientation were performed in the superconducting quantum interference device (SQUID) magnetometer (Quantum Design) in the temperature range 5–300 K and magnetic fields up to 5 T. The Quantum Design PPMS measuring system equipped with a 9 T magnet was used in few cases.

III. RESULTS AND DISCUSSION

A. Crystal structure

Since both UPdSi and UNiSi hydrides were synthesized for the first time, the determination of their crystal structure was the primary goal.

Structural properties of both materials are analogous and differ in lattice parameters only. Therefore, the structure modification upon hydrogen absorption will be illustrated in detail for the case of only one of them: UPdSi. It is important to stress that the following discussion concerns only the arrangement of metal atoms in the U*T*Si hydrides. The hydrogen atoms practically do not contribute to the intensities of the diffraction peaks due to the very small scattering factor. The x-ray powder diffraction pattern obtained for $UPdSiH_x$ has several interesting features: it shows notably less peaks than the pattern of the parent UPdSi [compare Figs. 1(a) and 1(b)], and the quality of the pattern (i.e., peak widths, background level) is unexpectedly good for the hydride. The reduction of the number of the diffraction peaks indicates that the structure of the hydride has a higher symmetry than that of pure UPdSi, which crystallizes in the orthorhombic TiNiSi-type structure [Fig. 2(a)].[10] This structure, as well as several others, can be derived by displacement-disorder transformations from the hexagonal AlB_2 unit cell shown in Fig. 2(b).

So it was natural to expect that the $UPdSiH_x$ lattice is one of the AlB_2 derivatives, too. A comprehensive description of the structure types relevant for hydrides belonging to this family is given, for example, in Ref. 14. The best matches to the experimental diffraction pattern were obtained assuming that $UPdSiH_x$ adopts either ZrBeSi- or GaGeLi-type hexagonal structure. The ZrBeSi structure type [Fig. 2(c)] can be obtained from the AlB_2 type [Fig. 2(a)] when its 2*d* site (1/3, 2/3, 1/2) is occupied selectively by Pd and Si atoms, so that the positions (1/3, 2/3, 1/2) and (2/3, 1/3, 1/2) are no longer equivalent. In addition Pd and Si atoms should be ordered along the *c* axis, forming the Pd-Si-Pd-Si-$\cdots$ chains. As the result the *c* parameter would be doubled compared to that of the AlB_2-type structure, and the *z* coordinate of Pd or Si would become $z=1/4$. In order to get the GaGeLi-type structure one more transformation should be added; namely, Pd and Si atoms should be moved off the plane $z=1/4$ in opposite directions. The assumption about doubling along the *c* axis is easy to check: if true, there should exist at least one reflection with an odd *l* parameter in a pattern indexed with the doubled *c*. On the contrary, if only even *l* indices are present, then the real parameter *c* is twice smaller than assumed. The diffraction pattern of $UPdSiH_x$ shows the strong (101) reflection as well as several other lines with an odd index *l*: namely, (103), (201), and (203). Consequently *c* is really close to 800 pm (Table I), and the crystal structure is either of ZrBeSi or GaGeLi type. Still the calculated pattern based on the GaGeLi structure model significantly deviates from the experimental intensities at high diffraction angles. This is not the case for the ZrBeSi-type structure, which provides the calculated pattern closely tracing the observed peak intensities. Therefore, the Rietveld analysis assuming that the UPdSi hydride has a hexagonal unit cell of the ZrBeSi type yielded a significantly lower value of the Bragg *R* factor compared to the one based on the GaGeLi-type structure model—namely, 11.7% vs 39.8%. X-ray diffraction showed also the presence of a small amount of UPdSi in the hydride. A summary of the fit is presented in Table I. Please note that due to the symmetry change upon hydrogenation, the values of the unit cell volume in the table are normalized per formula unit.

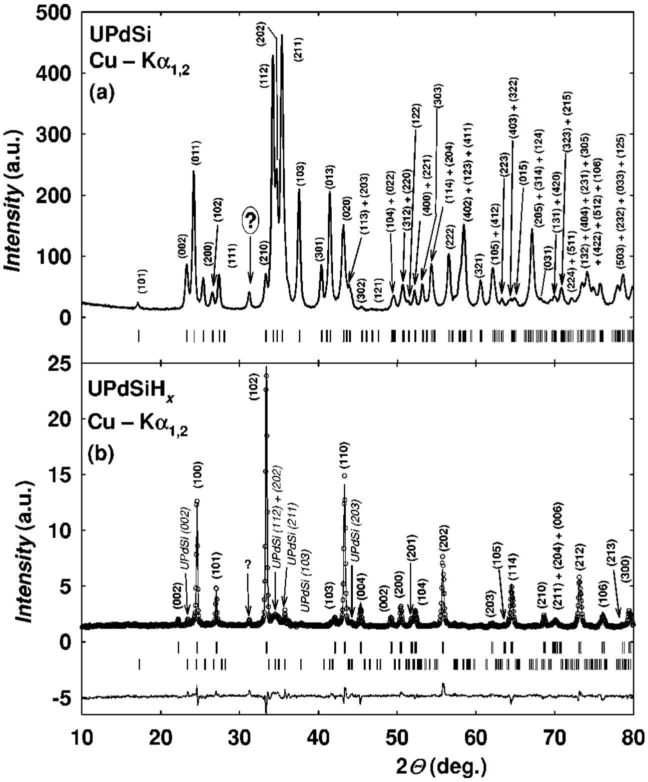

FIG. 1. (a) X-ray diffraction pattern of UPdSi. All peaks, except for the one marked by a question mark, are indexed within TiNiSi structure type. The calculated peak positions are shown by markers. (b) Experimental and calculated x-ray diffraction patterns for the UPdSi hydride as well as the difference curve. The upper row of markers denotes the positions of the $UPdSiH_x$ peaks and the lower one of the UPdSi peaks.

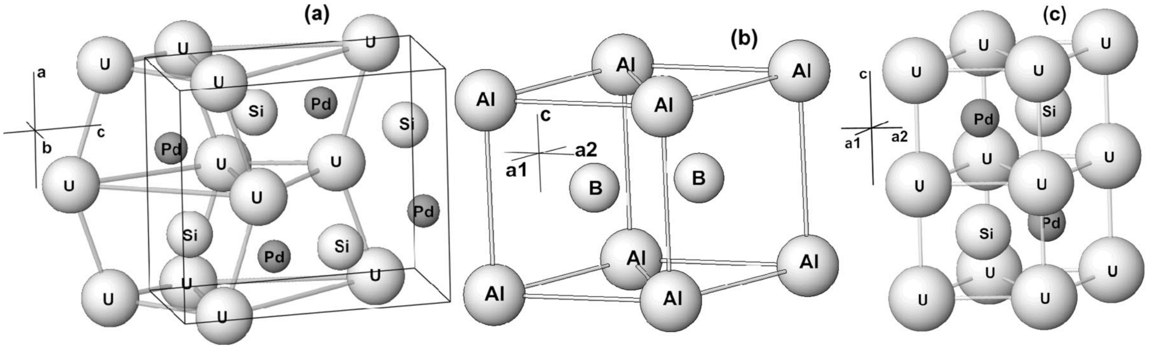

FIG. 2. (a) The unit cell of UPdSi-TiNiSi-type structure. The atoms outside the unit cell are shown to emphasize the similarity with the AlB_2-type structure. (b) Unit cell of the AlB_2-type crystal structure. (c) Unit cell of $UPdSiH_x$-ZrBeSi type. Note that crystal axes are interchanged as the symmetry gets modified: ***a***, ***b***, ***c*** of the AlB_2- or ZrBeSi-type structure correspond to ***b***, ***c***, ***a*** of the TiNiSi-type structure.

The conclusion is that $UPdSiH_x$ has the ZrBeSi-type [Fig. 2(c)] crystal structure and space group $P6_3/mmc$ (No. 194). The metal atoms occupy the following positions in the unit cell: U (0, 0, 0); Pd, Ni (1/3, 2/3, 1/4); Si (1/3, 2/3, 3/4) (positions of the H atoms have not been determined), which consists of alternating U and ordered Pd-Si layers stacked along the $\langle 001 \rangle$ direction and separated by exactly 1/4 of the parameter c. This arrangement transforms into the AlB_2-type structure if only one sort of atom occupies the Pd-Si layers or if both sites in this layer are occupied statistically by the atoms of different sorts. The latter can be indeed found in the U-Pd-Si and other U-T-Si systems with nonequal concentrations of T and Si atoms, usually described as as U_2TSi_3 compounds.[13]

The diffraction patterns of UNiSi and $UNiSiH_x$ show analogous variations as those of UPdSi-H. Also the ZrBeSi-type structure provides the best model for the Rietveld refinement of the x-ray data in the case of $UNiSiH_x$. The lowest value of the R factor achieved in this case was 19.5%. The structural data for the UNiSi hydride are listed in Table I.

The increase of symmetry upon hydrogenation is one of the two typical scenarios observed so far in the ternary ATX compounds ($A = f$-electron metal, T=transition metal, $X = p$-electron element) with the TiNiSi-type structure. It was reported, for example, for $TbNiSiD_{1.78}$,[14] $CeIrGaH_x$,[15] and $CeIrAlH_x$.[16] On the contrary, in NdNiSn-D deuteration results in the decrease of the lattice symmetry from the space group $Pnma$ to $Pna2_1$.[17] Neutron diffraction studies performed by Brinks and co-workers[14] on one of the compounds, which yields hexagonal symmetry after deuteration, $TbNiSiD_{1.78}$, give an idea about the hydrogen location in such structures. Namely, in the studied deuteride D atoms occupy the crystallographic position $4f$ (1/3, 2/3, z)—the interstitial site having 3 U atoms and 1 Pd atom as the nearest neighbors. Full occupation of this site would yield 2 H atoms/f.u. Our decomposition studies indicate that there are probably two hydrogen sites in $UTSiH_x$ because hydrides lose about 1 H atom/f.u. already at room temperature and then the temperature dependence of H_2 pressure shows a large step around 450 K followed by another one at 873 K. The decomposition of $UPdSiH_x$ and $UNiSiH_x$ by annealing in vacuum recovers the original orthorhombic crystal structure. The temperature of 923 K is required for the complete desorbtion of hydrogen. Diffraction patterns of the decomposed materials indeed match those of respective parent intermetallics.

B. Magnetic properties

1. UPdSi

Hydrogenation has a significant impact on magnetic properties of UPdSi. It increases the ordering temperature from $T_N = 31$ K in UPdSi to 46 K in the hydride, which was determined as the position of the maximum on the $M/H(T)$ curve [Figs. 3(a) and 3(b)].

TABLE I. Summary of crystal structure parameters (lattice parameters, interuranium spacing $d_{U\text{-}U}$, volume per formula unit, and its relative change) for UPdSi, UNiSi, and their hydrides.

	UPdSi	$UPdSiH_{1.0}$	UNiSi	$UNiSiH_{1.0}$
Structure	Orthorhombic (TiNiSi-type)	Hexagonal (ZrBeSi-type)	Orthorhombic (TiNiSi-type)	Hexagonal (ZrBeSi-type)
a (pm)	702.6	417.66(4)	695.2	403.2(1)
b (pm)	420.5	-	412.7	-
c (pm)	767.0	799.53(9)	705.5	777.6(2)
$d_{U\text{-}U}$ (pm)	348	417	329	403
Volume per f.u. (nm^3)	0.056 65	0.060 39	0.050 60	0.054 74
$\Delta V/V$ (%)	-	+ 6.6	-	+ 8.2

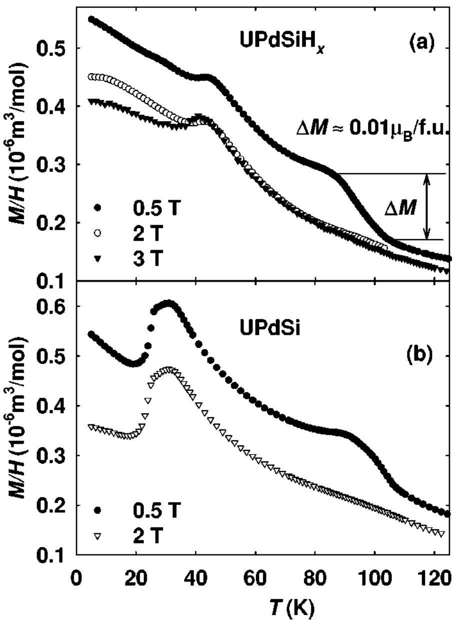

FIG. 3. Comparison of magnetic susceptibility curves for UPdSiH$_x$ (a) and UPdSi (b). The stepwise increase ΔM is due to an impurity phase.

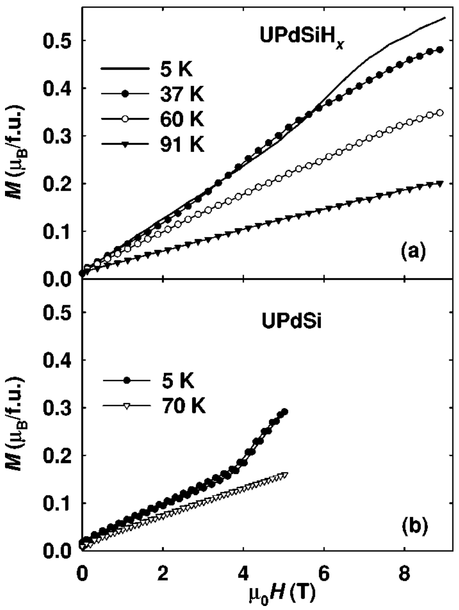

FIG. 4. The effect of hydrogenation on the field dependence of magnetization of UPdSiH$_x$. The solid lines are guides to the eye except for the one showing the $M(H)$ dependence at $T=5$ K in the hydride.

We attribute all other anomalies on the temperature dependence of susceptibility, i.e., the low-T upturn and the stepwise increase around 90 K, to the presence of UPdSi and another impurity phase, respectively. Indeed the low-T feature, which is especially well pronounced for $\mu_0H<0.5$ T, coincides with the magnetic phase transition temperature T_1 of the parent compound.[6,9] There is a certain ambiguity in assignment of the steep increase at about 90 K. From Fig. 3(a) it follows that this magnetic phase transition persists until $\mu_0H=0.5$ T, and it is suppressed at 2 T. Its extrinsic origin is deduced from the fact that the same anomaly is present in the original UPdSi [Fig. 3(b)] and even survives the decomposition: the magnetic susceptibility of the decomposed UPdSiH$_x$ shows the same steep increase. Thus, the 90 K anomaly has to be attributed to a secondary phase present already in the parent UPdSi. It may be, for example, off-stoichiometric UPd$_2$Si$_2$. UPd$_2$Si$_2$ normally orders antiferromagnetically at $T_N=136$ K,[18,19] but since in this case it forms as a side product of the UPdSi synthesis, the final spurious phase need not be of the exact 1:2:2 stoichiometry. Therefore, the resulting compound may not have fully compensated spins. This would explain the $M/H(T)$ values of UPdSiH$_x$ below 90 K, which are still too low for a ferromagnet, even though the transition itself reminds ferromagnetic rather than antiferromagnetic ordering. The magnetization increase at this transition is $\Delta M \approx 0.01\mu_B$/f.u. [Fig. 3(a)] and it fairly well corresponds to the remanent magnetization at 5 K [Figs. 4(a) and 4(b)].

All these considerations support the conclusion that UPdSiH$_x$ undergoes one magnetic phase transition at $T_{ord}=46$ K, which has an antiferromagnetic character.

The transition shifts to lower temperatures with increasing magnetic field, reaching 41 K at $\mu_0H=3$ T. This transition persists at least up to $\mu_0H=6$ T, the highest field applied during the susceptibility measurements. Fitting the temperature dependence measured in 6 T to the Curie-Weiss law [Fig. 5(a)] yields $\mu_{eff}=3.1\mu_B$/f.u. and $\Theta_P=-10$ K.

The value of the effective moment is typical for a higher degree of localization of the uranium $5f$ states (assuming that Pd carries no magnetic moment). The S-shaped magnetization curve shows that UPdSiH$_x$ undergoes a metamagnetic transition at $\mu_0H_c=6.3$ T at $T=5$ K [see Fig. 4(a)]. The critical field H_c decreases with increasing temperature and reaches 4 T at $T=37$ K. Above this temperature the

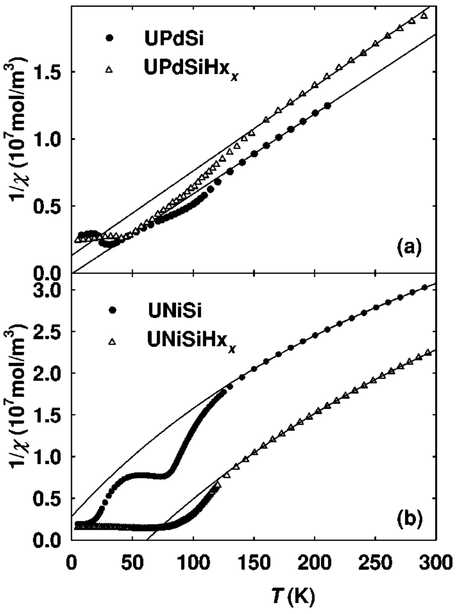

FIG. 5. Inverse susceptibilities of UPdSi-H (a) and UNiSi-H (b) systems. Solid lines indicate the results of the Curie-Weiss fits.

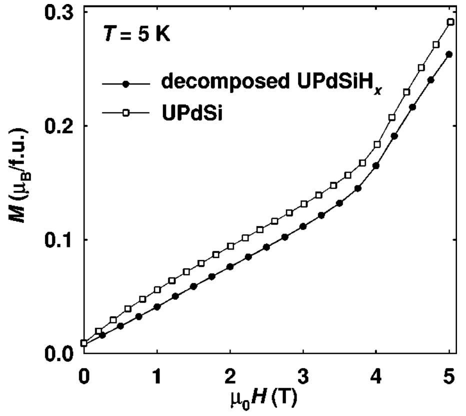

FIG. 6. Recovery of the original magnetic properties in fully decomposed UPdSi hydrides. Lines are guides to the eye.

metamagnetic transition vanishes. Since the transition persists at least up to 37 K, it should be related to the UPdSiH$_x$ phase and not to UPdSi, since the highest estimate of the T_N for the parent UPdSi is only 33 K according to Ref. 9 or 31 K according to our results [Fig. 3(b)]. This conclusion is corroborated by a comparison of the magnetization behavior of UPdSi and UPdSiH$_x$ [Figs. 4(a) and 4(b)], which show different critical fields: $\mu_0H_c = 4.2$ T in the parent compound and 6.2 T in the hydride. As demonstrated in Fig. 6, the decomposition of UPdSiH$_x$ recovers the original magnetic properties.

2. UNiSi

The magnetic properties of UNiSi are affected by hydrogenation to a larger extent than for UPdSi. As seen from Fig. 7(a), UNiSiH$_x$ exhibits three magnetic phase transitions at about $T_1 = 95$ K, $T_2 = 84$ K, and $T_3 = 61$ K, whereas for the parent UNiSi we have found, in agreement with Refs. 6 and 8, the following magnetic ordering temperatures: $T_N = 85$ K, $T_1 = 20$ K, and $T_2 = 10.5$ K [Fig. 7(b)]. The phase transitions at T_1 and T_2 in the hydride can be well distinguished only at very low fields [see Fig. 7(a)]. Above $\mu_0H = 0.5$ T they broaden so much that only two steps on the $M/H(T)$ curve indicate their presence. Above this field even the derivative dM/dT shows one broad peak in the place of previously existing two. If the magnetic field increases to $\mu_0H = 2$ T, only the transitions at T_2 and T_3 can be observed in UNiSiH$_x$: the value of T_2 does not change significantly compared to 0.5 T, while T_3 decreases to 60 K. In the field $\mu_0H = 5$ T, T_2 reaches 90 K, and $T_3 = 56$ K.

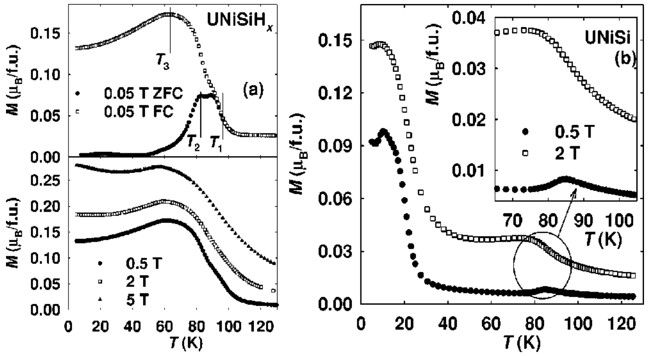

FIG. 7. a) Temperature dependence of the magnetization of UNiSi hydrides. The upper graph shows the low-field details of the transitions. Markers indicate the location of the critical temperatures T_1, T_2, and T_3 mentioned in the text. (b) $M/H(T)$ curves for UNiSi, measured in different magnetic fields. The inset shows the details of the antiferromagnetic phase transition in UNiSi.

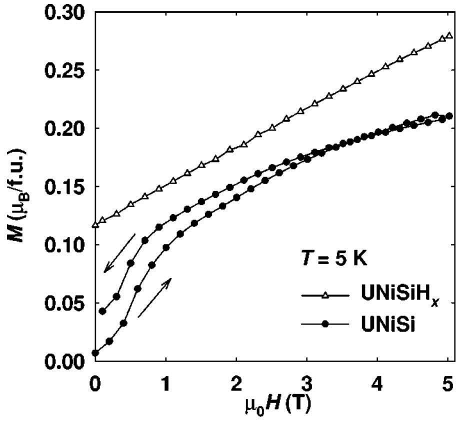

FIG. 8. The variations of the M vs H dependence in UNiSi due to hydrogenation. Lines are guides to the eye.

Magnetization measurements were performed in order to determine the type of ordering in the characteristic temperature ranges. It follows from Fig. 8 that the magnetization of UNiSiH$_x$ has a nonzero value at $H = 0$ ($T = 5$ K) and is almost linearly increasing with increasing magnetic field. The character of magnetization curves for higher temperatures is similar to that shown in Fig. 8, only the remanent magnetization decreases with increasing T and persists at least up until 92 K.

The Arrott plots indicate that the critical temperature of UNiSiH$_x$ is $T_C = 98$ K. Hence, based on the magnetic measurements we assume that the ground state of UNiSiH$_x$ is ferrimagnetic or ferromagnetic with $T_C = 98$ K, followed by two phase transitions at $T_1 = 84$ K and $T_2 = 61$ K. The broadening of the transitions at T_C and T_1 is the reason for which they cannot be distinguished at higher fields. Unlike UPdSiH$_x$, the paramagnetic susceptibility of UNiSiH$_x$ cannot be accounted for by the Curie-Weiss law. Application of the modified Curie-Weiss law [Fig. 5(b)] yields the following parameters: $\mu_{eff} = 2.3\mu_B$/f.u., $\Theta_P = 53$ K, and $\chi_0 = 1 \times 10^{-8}$ m^3/mol. The value of the effective moment of the hydride is higher than that of parent UNiSi ($\mu_{eff} = 1.9\mu_B$/f.u., $\Theta_P = -18$ K, and $\chi_0 = 1.4 \times 10^{-8}$ m^3/mol). The increase of the effective moment upon hydrogenation may be attributed to increased localization of the uranium $5f$ states. The higher and positive paramagnetic Curie temperature of the hydride demonstrates stronger ferromagnetic exchange interactions. The hydrogenation of UNiSi is reversible similarly to the case of UPdSi: UNiSiH$_x$ was decomposed by heating above $T = 923$ K, and magnetic properties of the resulting material were studied.

Figure 9 shows the comparison the $M/H(T)$ curves of the parent UNiSi and the product of decomposition. The similar-

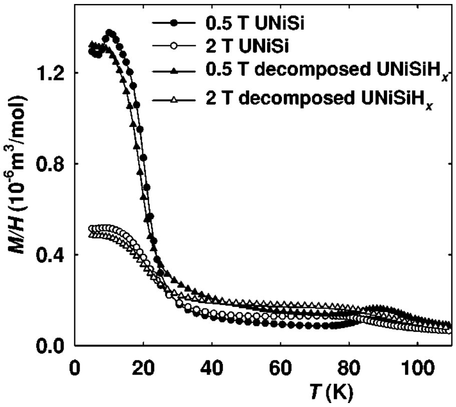

FIG. 9. Temperature dependence of magnetic susceptibilities of the parent UNiSi compound and UNiSi obtained by complete hydrogen desorbtion. Lines are guides to the eye.

ity of the curve shape and magnitude, and especially the critical temperatures, is apparent. The slight discrepancy in the absolute values and curve details may be attributed to the very fine-grain character of the decomposed samples, coming from decrepitation during the hydrogenation process. This also would explain that the features of its $M/H(T)$ dependences are more smooth.

Even though no neutron data are currently available for UPdSiH$_x$ and UNiSiH$_x$, some information about their magnetic structure can be deduced from the geometry of the unit cell. It follows from Fig. 2(c) that each U atom has nearest uranium neighbors either in the basal plane or along the c axis. In the first case the $d_{U\text{-}U}$ is equal to the parameter a of the unit cell and in the second case to $c/2$. The data from the Table I indicate that for both compounds $a > c/2$. In accordance with the empirical rule, the magnetic moments of uranium atoms tend to be perpendicular to shortest U-U link, i.e., to $\langle 001 \rangle$ for the studied materials. Therefore, we may expect the easy magnetization direction in UPdSiH$_x$ and UNiSiH$_x$ to be in the basal plane.

IV. CONCLUSIONS

UPdSi and UNiSi absorb hydrogen at a rather high temperature and pressure. The as-synthesized compounds contain about 2 H atoms/f.u., but the stable composition is approximately UTSiH$_{1.0}$. Incorporation of hydrogen into the crystal lattice leads to the expansion of the unit cell by 7%–8% and the symmetry changes from orthorhombic (TiNiSi type) in the parent compound to the hexagonal of the ZrBeSi type in the hydride. The original structure is restored when hydrogen is removed from the lattice by heating above $T = 923$ K. Another result of hydrogenation is the strengthening of magnetic exchange interactions in both UPdSiH$_x$ and UNiSiH$_x$ compared to nonhydrogenated samples.

UPdSi hydride orders at $T_N = 46$ K compared to 31 K in UPdSi and exhibits a metamagnetic transition at $\mu_0 H_c = 6.3$ T ($T = 5$ K) while the parent compound has $\mu_0 H_{c_{1,2}} = 4$ T and 7 T.[9] In UNiSi hydrogenation modifies also the type of magnetic order. The parent compound undergoes an antiferromagnetic transition at $T_N = 85$ K, while the hydride exhibits a ferromagnetic component and undergoes ordering at $T_C = 98$ K. The development of magnetic properties can be at least partly associated with enhanced localization of the $5f$ electronic states in hydrides due to enhanced U-U spacing. It reaches about 400 pm in both UPdSi and UNiSi hydrides. The influence of hydrogenation is more pronounced for UNiSi, where the original $d_{U\text{-}U}$ is lower than in UPdSi, while the final value for both hydrides is about the same.

ACKNOWLEDGMENTS

This work is a part of the research program No. MSM113200002, which is financed by the Ministry of Education of the Czech Republic, and it was also supported by the Grant Agency of the Czech Republic under Grant No. 202/01/P092.

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