

Hydride phases, structure, and magnetic properties of the UNiAlH_y system

P. Raj,* K. Shashikala, and A. Sathyamoorthy

Novel Materials and Structural Chemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai-400 085, India

N. Harish Kumar, C. R. Venkateswara Rao, and S. K. Malik

Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai-400 005, India

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The hydriding behavior of the heavy fermion compound UNiAl has been investigated. A range of hydride compositions with orthorhombic structure (space group $Pnma$) has been stabilized. As a result, a different nomenclature for the UNiAlH_y hydride phases is proposed. X-ray diffraction and magnetic studies are reported on three compositions in this range of hydrides, viz., $y=0.06, 0.14,$ and 0.58 . All these compositions do not show magnetic ordering down to 2 K, though weak ferromagnetic correlations are apparent in their magnetic studies. Low effective moment values on uranium atoms in these cases, appreciably below the value for itinerant antiferromagnetic UNiAl, indicate increased itinerant character of U $5f$ electrons. We also report magnetization studies on hexagonal UNiAlH_y ($y=0.7$). It shows ferromagnetic ordering below 87 K. This means that in the hexagonal hydride phases, the nature of magnetic ordering changes from antiferromagnetic ($y=0$) to ferromagnetic ($y=0.7$) and again back to antiferromagnetic for $y=2.3$. It is shown that the moment on the uranium atoms in the hexagonal hydride phases ($y\cong 0.7$ and $2.0-2.5$) is close to the free-ion local moment value.

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I. INTRODUCTION

Uranium-based intermetallic compounds present a highly fascinating area of scientific investigations because of the wide variety of anomalous phenomena exhibited by them. The unusual properties of these compounds are related to the hybridization between U $5f$ electrons and the electrons on the surrounding atoms. The U $5f$ electrons lie on the borderline between localized and itinerant behavior and are therefore susceptible to undergo a change in the degree of (de)localization, depending on the strength of hybridization. The development of the magnetic properties from the temperature-independent Pauli paramagnetic state (i.e., itinerant and in some cases even superconducting at the lowest temperatures) to spin fluctuation and finally to the local moment behavior also follows the extent of hybridization.¹⁻⁷ This makes the crystallographic as well as electronic structure of these intermetallic compounds quite sensitive to external perturbations like temperature, pressure, magnetic field, and changes in the local surroundings of U atoms.^{6,7} Hydrogen absorption can also be gainfully employed to probe the structural and magnetic instabilities of the U-based intermetallic compounds. In fact, it offers the advantage, at least in cases where H₂ absorption does not cause structural change, of retaining the surrounding metallic neighbors of U atoms intact.^{3,7} It may, however, be noted that very few studies of this kind have been reported for uranium-based systems. This is basically because, in most uranium containing systems, H₂ absorption does not take place. In most cases, it has been found to result in disproportionation, forming binary hydride (UH₃) plus some uranium-deficient compound, instead of ternary (or quaternary) compound hydride.⁸

UNiAl is one among the few compounds which show hydrogen (deuterium) absorption without disproportionation.^{5,7-13} It is reported to form two hydride (deuteride)

phases, wherein each phase retains the hexagonal (Fe₂P-type or more accurately the HoNiAl-type, space group $P\bar{6}2m$) structure of the parent compound, which is considered unusual.⁸ As regards the composition of the two UNiAlH_y hydride phases, the lower hydride/deuteride phase is reported to have $y\cong 0.7-0.8$.^{5,8} The value of y for the higher hydride phase is, however, reported to be quite at variance, being 1.9, 2.2,^{8,10} 2.5,¹³ and 2.7.¹⁴ According to our knowledge, there are only two reports on the magnetization measurements on UNiAlH_y system.^{9,10} The studies by Zogal *et al.*⁹ pertain to an inadvertently formed biphasic hydride system. Their hydride sample, having an average value of $y=1.9$, showed two antiferromagnetic transitions with T_N values of 122 and 34 K. The latter transition was ascribed possibly to the hydrogen-solubility composition. In a very recent work, Kolomiets *et al.*¹⁰ have reported antiferromagnetic ordering for UNiAlH_{2.3} and UNiAlD_{2.1} with $T_N=99$ and 94 K, respectively. There are no reports of magnetization studies in the lower hydride phase with $y=0.7$.

We have very recently reported hydriding behavior, Mössbauer, and magnetic studies of the U(Fe_{1-x}Ni_x)Al system for $0\leq x\leq 0.75$.⁷ It was found that H₂ absorption does not take place for $x<0.7$ and that the maximum value of y in U(Fe_{0.3}Ni_{0.7})AlH_y was $\cong 0.8$, which matches almost exactly with the hexagonal hydride phase of UNiAl, with similar H content. It was also found that no other hydride composition stabilizes either along the absorption or the desorption route. Further, from the ⁵⁷Fe Mössbauer studies, it was inferred that the electron charge transfer from hydrogen to the Fe $3d$ band (implying possibly to the Ni $3d$ band also) results in a reduction of the $5f-3d$ hybridization, which in turn enhances the ferromagnetic correlations. Thus the large increase in ferromagnetic Curie temperature (T_C), paramagnetic Curie temperature (θ_P), and effective magnetic moment (μ_{eff}) val-

TABLE I. Structural and magnetic parameters of the UNiAlH_y system.

y	Structural parameters							Magnetic parameters				
	Structure type	Cell parameters (Å)	Position parameters			Cell vol./ f.u. (Å ³)	Min U-U distance (Å)	Temperature range (K)	Θ _p (K)	χ ₀ (10 ⁻³ emu/mol)	μ _{eff} (μ _B /f.u.)	Mag. at H _{app} =55 kOe (μ _B /f.u.)
0.0	Hexagonal	a = 6.7347 c = 4.0371	x 0.579		0.198	52.86	3.49	30–300 150–300	-27.7 -26.9	1.03 1.03	1.63 1.62	
0.06	Orthorhombic	a = 7.058 b = 3.920 c = 7.434	x 0.517	0.143	0.765	51.42	2.97					
0.14	Orthorhombic	a = 7.008 b = 3.934 c = 7.470	x 0.509	0.129	0.772	51.49	3.01	30–300 150–300	1.2 5.2	1.35 1.37	0.53 0.50	0.03
0.58	Orthorhombic	a = 6.722 b = 4.130 c = 7.904	x 0.513	0.210	0.776	54.86	3.69	30–300 150–300	8.0 -117.0	1.57 0.46	1.29 2.38	0.28
0.70	Hexagonal	a = 6.9909 c = 3.9910	x 0.590		0.256	56.31	3.66	90–300 150–300	84.1 82.4	0.44 0.31	2.90 2.94	1.38
2.30	Hexagonal	a = 7.1844 c = 4.0244	x 0.668		0.322	59.96	4.16	110–300	-96.1	0.36	3.02	

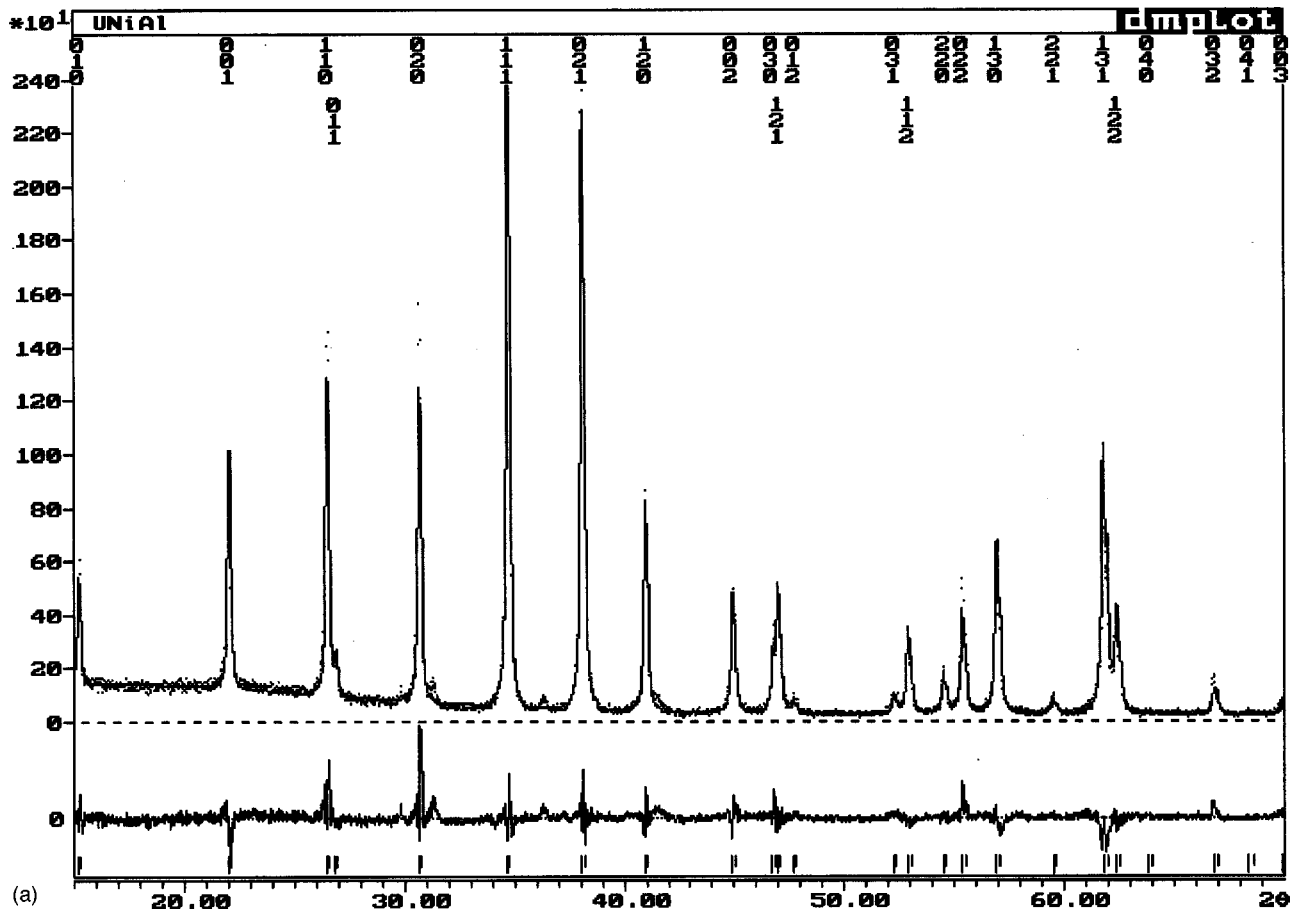


FIG. 1. (a), (b), (c), (d), and (e) X-ray diffraction patterns for UNiAlH_y with y=0, 0.14, 0.58, 0.7, and 2.3, respectively.

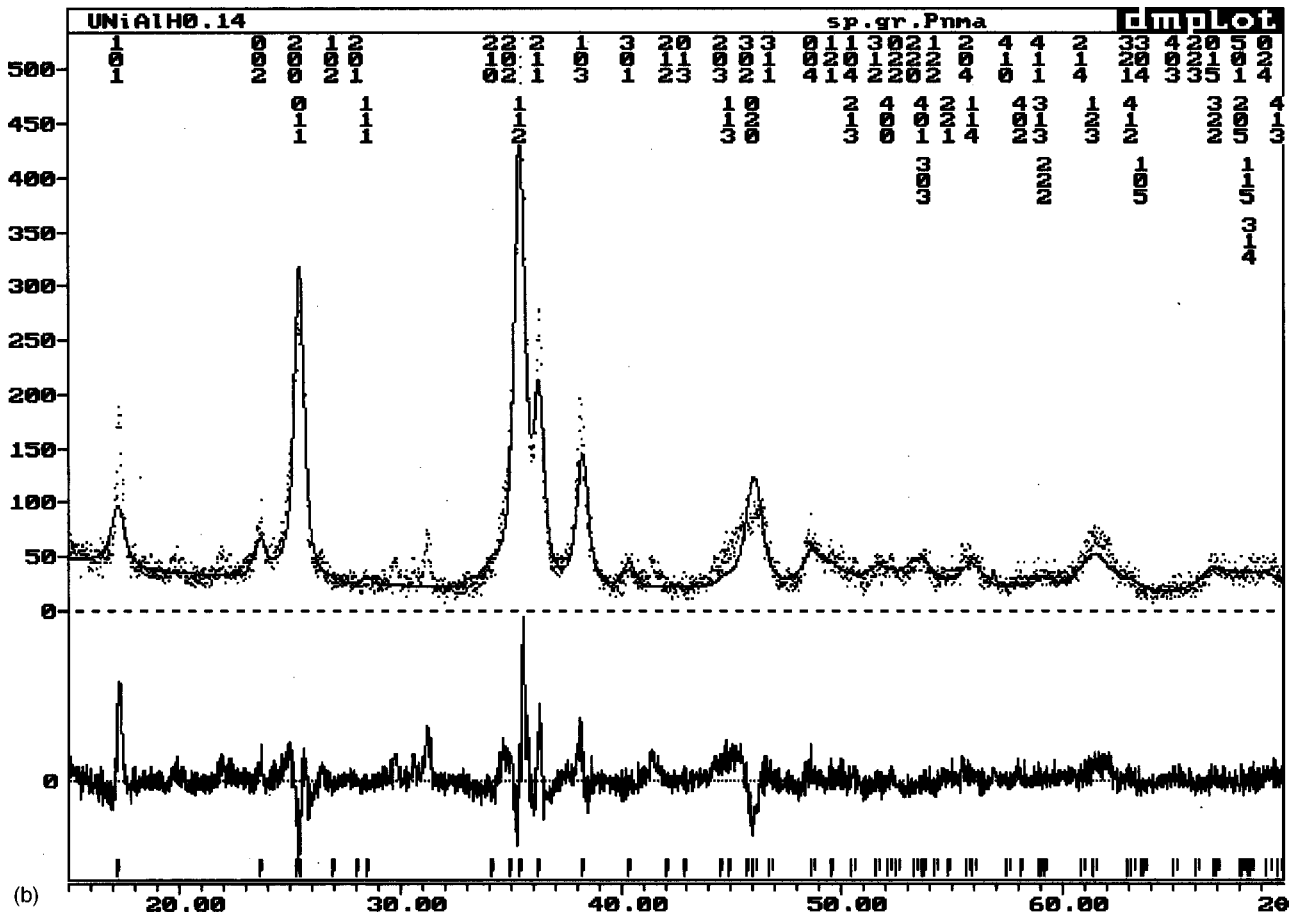


FIG. 1. (Continued).

ues could be explained. This also implied that even hexagonal UNiAlH_y ($y=0.7-0.8$) should also order ferromagnetically.

In the present investigation, we have carried out elaborate studies on the hydriding behavior of pure UNiAl along the absorption as well as desorption routes. It is found that a new hydride phase coexists with the $y \approx 2.3 \pm 0.2$ hydride phase. The same could be stabilized in almost single phase for values of y down to 0.06 along the desorption route. The new hydride phase is found to have different structure (orthorhombic) compared to that of the parent compound (hexagonal), even for $y \approx 0.06$. The structural as well as magnetic parameters of all the hydride phases in the UNiAl-H₂ system have been determined. It is shown that (i) the hexagonal hydride phase with $y=0.7$ orders ferromagnetically below $T_C=87$ K and (ii) the moment on the U atoms, in the hexagonal hydride phases, is close to the free-ion local moment value. In the orthorhombic hydride region, however, the itinerancy increases relative to parent itinerant antiferromagnetic compound UNiAl.

II. EXPERIMENT

UNiAl was prepared by argon arc melting the pure metals in stoichiometric proportion. Melting was done several times, and the button was turned upside down after every melting for better homogenization. Hydriding was carried

out following the standard procedure involving the activation of the compound, before introducing H₂ in the reaction chamber. For preparing the hydride with the highest H₂ content (y ranging from ≈ 2 to ≈ 2.5), a maximum of 45 atm of H₂ pressure was employed. The sample was cycled between room temperature and 473 K several times. In order to obtain hexagonal hydride with y varying from ≈ 0.7 to 0.8, in single-phase form, the maximum H₂ pressure was restricted to ≈ 0.7 atm, and similar temperature cycling was employed. In the case of incomplete conversion of the compound to the above-mentioned hydride phases, it was found that (i) the phase coexisting with the hexagonal hydride with $y \approx 0.7$ was the parent compound itself and (ii) the phase coexisting with the one with highest hydrogen content was a hitherto unknown phase. In order to prepare this unknown phase in its single-phase form, the hydride with maximum y value was heated at a temperature ≈ 463 K, while simultaneously evacuating the liberated hydrogen. Following this procedure, we have stabilized and studied three compositions with $y = 0.06, 0.14,$ and 0.58 . It may be mentioned that in this procedure, a prolonged evacuation can result in partial conversion to the parent compound. Also, a small distribution around the average y value can be expected because of the solubility range in this hydride phase. The procedure was standardized to minimize the presence of the parent compound. For further experiments, the hydrides formed in the

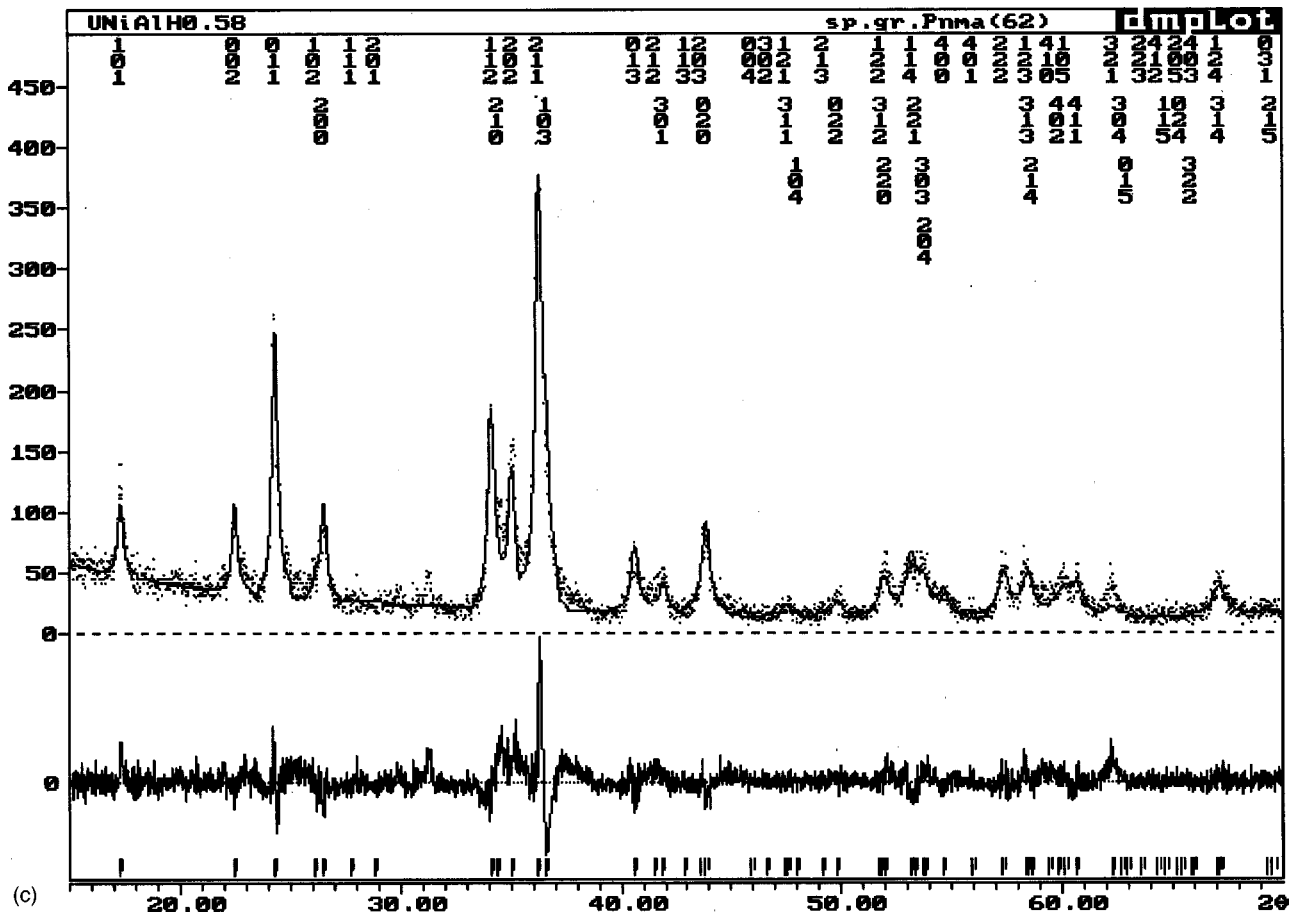


FIG. 1. (Continued).

reaction chamber were cooled to liquid nitrogen temperature before exposing them to the ambient atmosphere. The amount of hydrogen absorbed by a given sample was estimated by (i) monitoring the change in H_2 pressure in the absorption process, as well as by (ii) complete decomposition of a part of the hydride sample in an evacuated and calibrated chamber. Our estimates along the two routes agreed within the experimental error of less than $\pm 3\%$. The hydrogen content in samples with low values of $y \approx 0.1$ had a maximum uncertainty of $\pm 10\%$. X-ray diffraction studies were used to ascertain the single-phase formation and Rietveld profile analysis of the data was used to derive the structural parameters. Magnetization measurements, in the temperature range 2–300 K and in magnetic fields up to 55 kOe, were carried out using a commercial superconducting quantum interference device (SQUID) magnetometer (Quantum Design). The magnetization vs temperature data were fitted to a modified Curie-Weiss law to obtain θ_P , χ_0 , and μ_{eff} values.

III. RESULTS AND DISCUSSION

Intermetallic compounds with a general formula ATX (A = uranium or rare-earth element, $T = 3d, 4d,$ or $5d$ atom, and $X = p$ -electron atom like Si, Ge, etc.) are among the most widely studied systems. These compounds are reported to show systematic trends as a function of the electronic con-

figuration as well as the size of the constituent atoms. A large number of these intermetallic compounds stabilize in the hexagonal, Fe_2P -type or more accurately $HoNiAl$ -type structure, with space group $P\bar{6}2m$. $UNiAl$ belongs to this category. In this case, the structure consists of alternate planes containing Al and Ni metal atoms at $z=0$ and U and Ni atoms at $z=(1/2)$. The Ni atoms in these planes occupy 2(c) and 1(b) sites, respectively. The U and Al atoms occupy 3(g) and 3(h) sites, respectively. It is now widely accepted that the magnetic properties are mainly governed by hybridization between uranium and other constituent metal atoms. The contribution from direct U-U overlap towards the U-moment (de)stabilization is considered to be less important,¹ even when the separation between near-neighbor uranium atoms is less than the Hill limit. In $UNiAl$, U-U separation equals 3.49 Å (Table I), which is very close to the Hill limit. In such a situation, the hybridization due to direct overlap between the neighboring U atoms is reported to give a negligible contribution to the magnetic properties.¹ In the new $UNiAlH_y$ hydrides, however, it appears that, at least for $y=0.06$ and 0.14, the direct U-U hybridization may have appreciable role towards $5f$ -electron delocalization and consequent loss of uranium moment value. This is because the U-U separation, which is found to be ≈ 3 Å, is much smaller than the Hill limit. This is not the case for $y=0.58$ hydride, where the U-U separation (3.48 Å) is very similar to that of

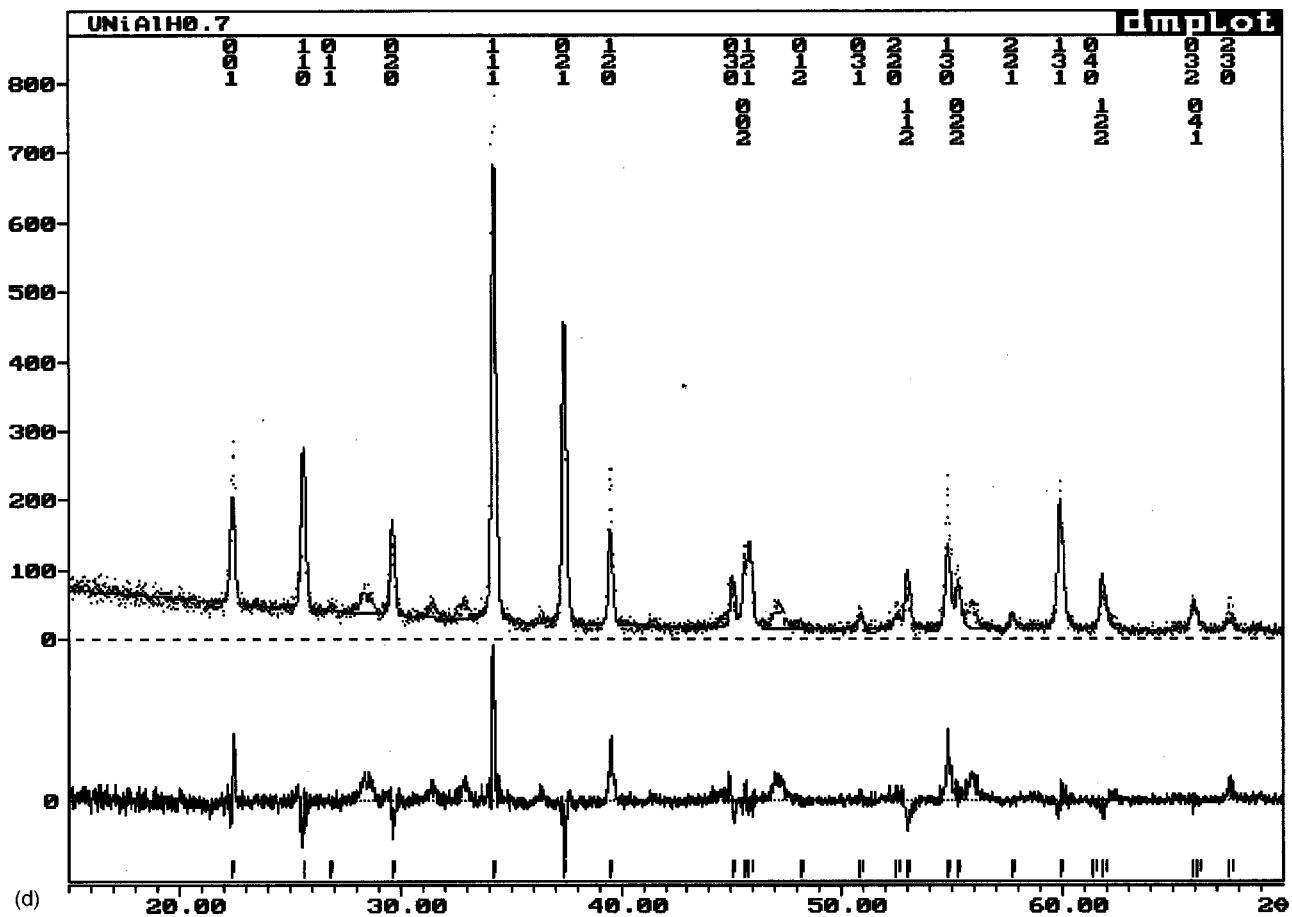


FIG. 1. (Continued).

the parent compound. In this case, the nonobservation of magnetic ordering down to 2 K can only be understood (qualitatively) in terms of structural differences relative to UNiAl. In Table I we have also listed the values of volume per formula unit. Based on the lower value of this volume for $y=0.06$ and 0.14 (relative to the parent compound) reduction in U-moment value may be expected, but not for $y=0.58$ case where this volume has increased.

A. Structure of various UNiAlH_y hydride phases

Figures 1(a), 1(b), 1(c), 1(d), and 1(e) show the x-ray diffraction patterns of UNiAlH_y, for $y=0, 0.14, 0.58, 0.70,$ and 2.3 , respectively, along with the results of Rietveld profile analysis. It may be noted that a small amount of impurity phase lines (most likely due to uranium suboxides) are also present. It will be shown latter that (i) the impurity phase does not show magnetic ordering down to 2 K and (ii) the moment associated with the impurity phase, if any, is very small, so that all the observed magnetic properties are characteristic of the main phase. The x-ray spectrum for $y=0.06$ looks very similar to that of 0.14 and has not been plotted. The fitted structural parameters are listed in Table I. It is seen that in a given structure type, the change in lattice parameters is highly anisotropic. For example, as y changes from 0.06 to 0.14 and to 0.58 , the lattice parameter a de-

creases from 7.058 to 7.008 and to 6.722 Å. The b and c cell parameters, on the other hand, increase with increasing hydrogen content. The corresponding values for the three compositions are found to be $b=3.920, 3.934,$ and 4.130 Å and $c=7.434, 7.470,$ and 7.904 Å, respectively. An important observation that needs to be emphasized here is that even a very small amount of hydrogen, $y\cong 0.06$ (i.e., hydrogen-to-metal ratio of 0.02), destabilizes the hexagonal structure of UNiAl and converts it to the orthorhombic form (TiNiSi type, space group $Pnma$ No. 62). This clearly brings out the built-in structural instability, which is typical of the actinide compounds where $5f$ electrons are on the borderline between itinerant and localized character. We will, hereafter, refer to the hydrides, with y values ranging from $\cong 0.06$ to $\cong 0.58$ and having orthorhombic structure, as the β phase. This low hydrogen containing phase has to be distinguished from the normally accepted α -hydride phase, wherein hydrogen dissolves in the parent compound, without change of structure. In a very recent study Yamanaka *et al.*¹⁵ have presented a nomenclature based on the already reported hydride phases. The maximum hydrogen content in their α -hydride phase corresponds to $y\leq 0.02$. In view of the hydride phase found in our present work, a nomenclature needs to be prescribed for the higher hydride phases. Therefore, the hexagonal hydrides with approximate composition $y\cong 0.7-0.8$ and $\cong 2.0-2.5$ will be termed as γ and δ phases, respectively.

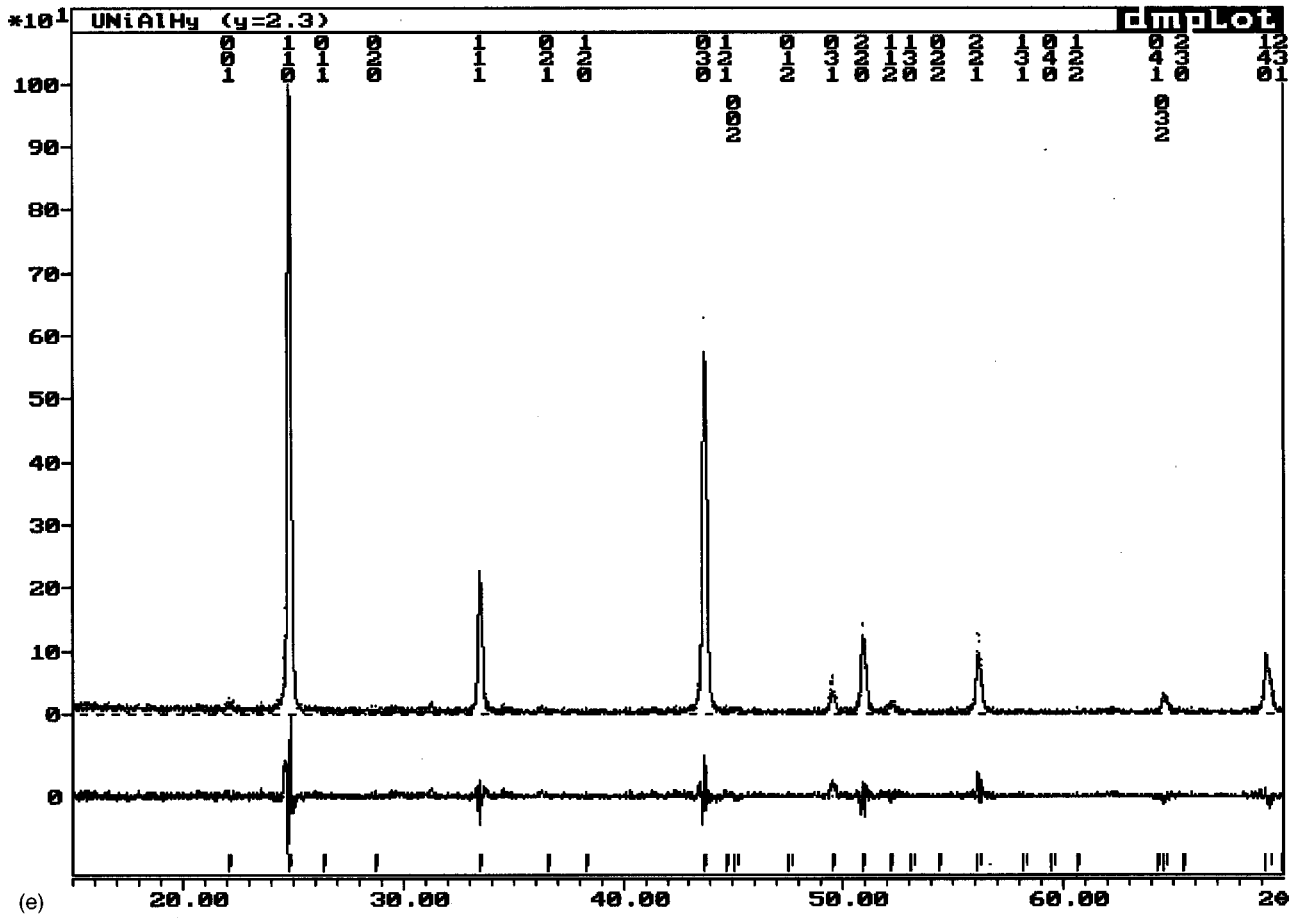


FIG. 1. (Continued).

The absorbed hydrogen brings about a considerable change in the magnetic properties of the compound, as described below.

B. Magnetic properties

UNiAl is a well-known heavy fermion compound and is reported to show itinerant antiferromagnetic (AF) ordering. The reported T_N values range from 18 K (Ref. 16) to 23 K.⁹ Figure 2 shows the temperature dependence of molar suscep-

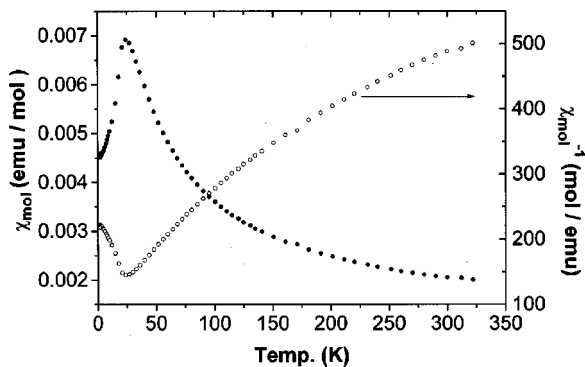


FIG. 2. Temperature dependence of χ and χ^{-1} for UNiAl ($H_{app} = 5$ kOe).

tibility (χ) for our polycrystalline UNiAl under an applied field (H_{app}) of 5 kOe. A clear peak at ≈ 25 K and the derived value of $T_N \approx 19$ K (not shown) are in complete agreement with Ref. 6. It may be noted that the T_N in UNiAl and many other antiferromagnetic systems is identified with the maximum in $\partial(\chi T)/\partial T$ and not with the maximum in susceptibility. The latter is $\approx 5-7$ K higher.⁶ The value of $\mu_{eff} \approx 1.6\mu_B/f.u.$ is appreciably below that expected for free-ion U^{3+} or U^{4+} values ($\approx 3.6\mu_B$). This low value is in conformity with partial delocalization of $U 5f$ electrons and heavy fermion behavior.

In Fig. 3(a), the temperature dependences of χ and χ^{-1} are shown for the $y = 0.14$ sample. A large increase in the χ value at low temperatures indicates the onset of ferromagnetic correlations at low temperatures. This is corroborated from the observed hysteresis curve at 5 K, shown in Fig. 3(b). The magnetization curve does show a slight bending towards the field axis, but shows no tendency to saturate up to 55 kOe, and the hysteresis curve shows zero remanent magnetization. Further, the value of magnetization for an applied field of 55 kOe is very small ($\sim 0.03\mu_B/f.u.$). This also implies that the contribution to magnetization arising from the impurity phase, if any, is negligibly small. From our field-dependent magnetization studies at various temperatures, we conclude that for this hydride composition, al-

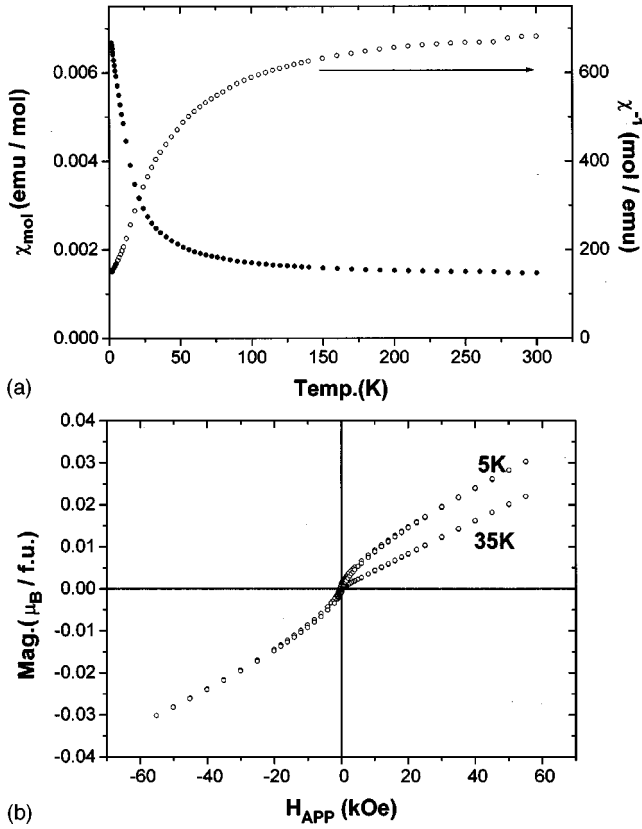


FIG. 3. (a) Temperature dependence of χ and χ^{-1} for $\text{UNiAlH}_{0.14}$ ($H_{\text{app}}=5$ kOe). (b) Magnetic hysteresis curve for $\text{UNiAlH}_{0.14}$ at 5 and 35 K.

though appreciable ferromagnetic correlations set in below ~ 10 K, long-range ferromagnetic ordering does not percolate into the bulk down to ≈ 5 K. A small positive value of $\theta_p \approx 1$ to 5 K and quite low value of $\mu_{\text{eff}} = 0.5\mu_B/\text{f.u.}$ (see Table I) also corroborate the presence of weak ferromagnetic correlations. Since the concentration of U atoms should be related mainly to the change of structure. From Table I, it is seen that for $y=0.06$ and 0.14 , the U-U separation is unusually small, ≈ 3 Å, which is appreciably below the Hill limit. In this case, U-U hybridization may also be expected to delocalize $5f$ electrons and consequent loss of U moment. Even in this case, the hybridization between U $5f$ electrons with those of the ligands is considered to be the major component towards delocalization.¹ The above observations imply that if UNiAl could be stabilized in orthorhombic structure, the moment on U atoms would be quite low.

Figure 4(a) shows the temperature dependence of χ and χ^{-1} for orthorhombic UNiAlH_y with highest hydrogen concentration ($y=0.58$) studied here. No magnetic ordering is indicated. The values of θ_p , χ_0 , and μ_{eff} derived from these data are listed in Table I. Unlike the case of $y=0.14$ sample, the values of these fitted parameters change very considerably when susceptibility data in a different range of temperatures are used to obtain a least-squares fitting. For example, the fitted value of θ_p from 150–300 K data is -117 K, whereas that obtained from 30–300 K data is $+8$ K. The values of other parameters are also similarly quite different.

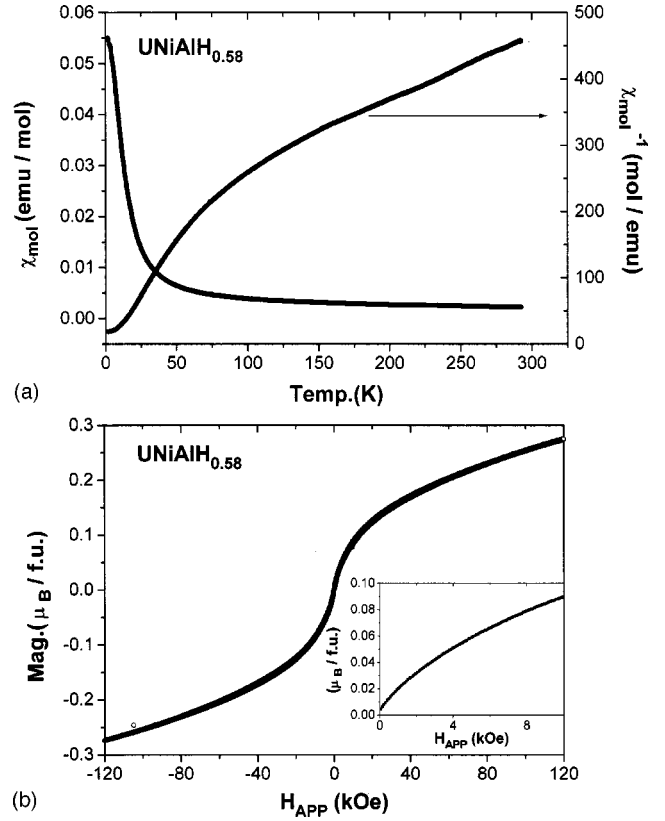


FIG. 4. (a) Temperature dependence of χ and χ^{-1} for $\text{UNiAlH}_{0.58}$ ($H_{\text{app}}=5$ kOe). (b) Magnetic hysteresis curve for $\text{UNiAlH}_{0.58}$ at 2 K.

This means that AF correlations prevalent in the high-temperature region change over to ferromagnetic type at low temperatures. All the same, the magnetic ordering does not take place down to 2 K. This is supported by the hysteresis curve at 2 K shown in Fig. 4(b). The presence of ferromagnetic correlations is seen in the form of bending of the curve towards the field axis, but the zero value of the remanent magnetization rules out ferromagnetic ordering out down to 2 K.

Figure 5(a) shows the temperature dependence of χ for hexagonal UNiAlH_y for $y=0.7$ ($H_{\text{app}}=5$ kOe), which is typical of a ferromagnetically ordered state. The ferromagnetic ordering temperature (T_C) is found to be ≈ 87 K. Large positive values of $\theta_p \approx 84$ K and $\mu_{\text{eff}} = 2.9\mu_B/\text{f.u.}$ are derived from the paramagnetic state data. The field dependence of magnetization, shown in Fig. 5(b), gives a saturation moment value of $\approx 1.4\mu_B/\text{f.u.}$, and the remanent magnetization is also close to this value. The above observations not only support ferromagnetic ordering for this γ -hydride phase, but also indicate increased local moment on U atoms relative to the orthorhombic hydride phase as well as unhydrided compound.

Figure 6 shows the temperature variation of χ and χ^{-1} for the $y=2.3$ hexagonal hydride phase. A peak in the χ vs temperature curve is found at ≈ 105 K. The value of T_N , as defined earlier, being the temperature having a maximum in $\partial(\chi T)/\partial T$, is found to be ≈ 98 K. The parameters derived from the paramagnetic region are found to be $\theta_p \approx -96$ K

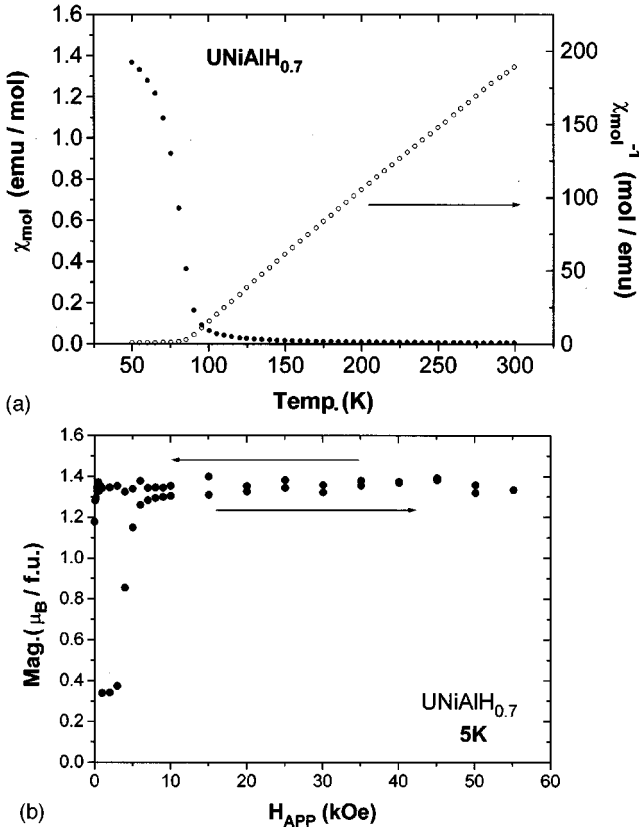


FIG. 5. (a) Temperature dependence of χ and χ^{-1} for $\text{UNiAlH}_{0.7}$ ($H_{\text{app}}=5$ kOe). (b) Field dependence of magnetization for $\text{UNiAlH}_{0.7}$ at 5 K (only the first quadrant).

and $\mu_{\text{eff}}=3.0\mu_B/\text{f.u.}$. This means that, as in the case of the $y=0.7$ composition, the uranium moment is fairly localized.

C. Magnetic properties and 5f-3d hybridization

We will now try to rationalize the observed variation in the magnetic properties, as a function of hydrogen content, in terms of hybridization. It should, however, be emphasized that only qualitative trends can be predicted. This is because the hydriding not only changes the transition-metal d -band occupancy, the lattice expansion is also anisotropic. In the present system, even structural changes are found. This results in a quite different variation of the competing hybridization components, viz., U-U, U-T, and U-X.

In hexagonal UTX compounds of the type UNiAl , it has been suggested that the magnetic interaction between U 5f electrons and d electrons of the T atoms is ferromagnetic in nature, whereas the interaction between the U 5f and p electrons of the X atoms is of antiferromagnetic type.¹⁷ As the number of electrons in the d shell increases (which is similar to the T atom moving to the right of the periodic table), the U-T hybridization decreases and ferromagnetic interaction gains in strength. We had explained a large increase in T_C , θ_P , and μ_{eff} values, on hydriding $\text{U}(\text{Fe}_{0.3}\text{Ni}_{0.7})\text{Al}$ to $\text{U}(\text{Fe}_{0.3}\text{Ni}_{0.7})\text{AlH}_{0.8}$, in terms of charge transfer from the H atom to the 3d band of transition metals.⁷ The filled 3d band results in a reduction of the hybridization between U 5f and

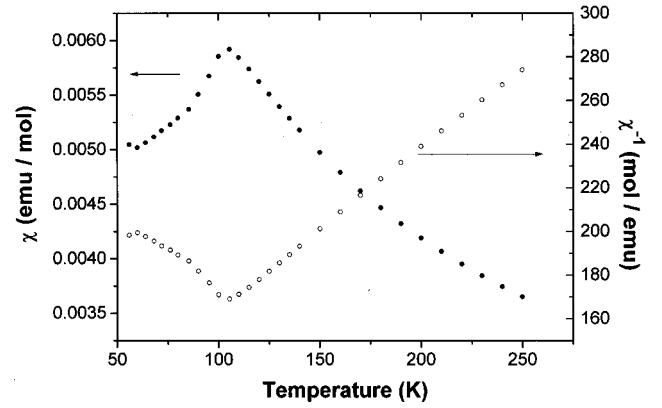


FIG. 6. Temperature dependence of χ and χ^{-1} for $\text{UNiAlH}_{2.3}$ ($H_{\text{app}}=5$ kOe).

Ni 3d electrons, thus strengthening the ferromagnetic correlations. Similar arguments can be directly extended to the γ -hydride phase of the present system. It may be noted that all the magnetic parameters, viz., T_C , θ_P , and μ_{eff} , for the γ -hydride phase (in Table I) are very similar to those reported earlier for $\text{U}(\text{Fe}_{0.3}\text{Ni}_{0.7})\text{AlH}_{0.8}$.⁷ Thus we infer that at least in the hexagonal structure of the HoNiAl -type, electron charge transfer from H to the 3d band plays an important role in determining the strength of ferromagnetic interaction.

The antiferromagnetic ordering of the δ -hydride phase ($y\approx 2.3$) cannot be understood by simply extending the argument of further increase in 3d-band occupancy. It can, however, be understood from the following arguments. Satisfactory Rietveld fitting of the x-ray diffraction data of the hexagonal hydride phase with $y=2.3$ requires that there be no Ni atoms in the plane containing U atoms. In fact, the Ni atoms which exist in the 1(b) position in UNiAl and the γ -hydride phase are shifted to the 1(a) position or Al-Ni plane in the δ -hydride phase, so that all Ni and Al atoms now lie in the same plane. The same conclusion has been drawn in recently reported neutron diffraction study of $\text{UNiAlD}_{2.2}$.⁸ These authors also reported that the D atom lies close to the U-atom plane. Complete absence of T atoms in the U plane drastically reduces the ferromagnetic interactions, so that the antiferromagnetic interactions between U and Al atoms can account for the observed antiferromagnetic ordering.

It appears that the explanation put forward for the hexagonal hydride phases cannot be directly extended to the orthorhombic structure. At present, we do not have a satisfactory explanation for the loss of uranium magnetic moment and nonobservance of magnetic ordering in the orthorhombic β -hydride phase. In fact, the reason for the very different magnetic properties of orthorhombic β - $\text{UNiAlH}_{0.58}$ and hexagonal γ - $\text{UNiAlH}_{0.7}$, both having similar values of H content as well as minimum U-U separation, has to be related to their structural differences. At present, this is not understood.

IV. CONCLUSIONS

It is shown that in the UNiAlH_y system, in addition to the reported hexagonal hydride phases with $y=0.7-0.8$ and

≈ 2 – 2.3 , a new range of hydride compositions with orthorhombic structure (space group $Pnma$) also exists. This necessitates a new nomenclature for all the hydride phases, the same has been proposed. Three composition with $y=0.06$, 0.14 , and 0.58 , belonging to the new orthorhombic structure, have been stabilized and their structural and magnetic parameters have been estimated. These compositions show an increased itinerancy of $5f$ electrons relative to that in the parent compound $UNiAl$ and do not show magnetic ordering down to 2 K. The reduction in the U-moment values, in compositions with $y=0.06$ and 0.14 , is likely to have a con-

tribution from hybridization between the neighboring U atoms also, which are unusually close (≈ 3 Å). Magnetization studies on hexagonal hydride phase, $UNiAlH_{0.7}$, are reported in this paper. In hexagonal hydride phases, the changeover from antiferromagnetic to ferromagnetic and again to the antiferromagnetic ordering, as y increases from 0 to 0.7 and to 2.3 , has been explained. The increase in ferromagnetic correlations and the localization of moment on U atoms has been rationalized in terms of increasing $3d$ -band occupancy with increasing H content.

*Author to whom correspondence should be addressed. Electronic address: praj@apsara.barc.ernet.in

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