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Neutron-diffraction studies of single crystals of U_2T_2In ($T=Ni$, Pd, Pt)

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Neutron-diffraction experiments on single crystals of U_2T_2In ($T=Ni$, Pd, Pt) verify the noncollinear magnetic structures found with studies of polycrystalline samples, and determine the values of the ordered moments. Compared to the earlier studies, we find the ordered moments on the uranium atoms larger, and no moment associated with the T atoms, in disagreement with one previous study. The $T=Pt$ sample has a large electronic specific heat and does not order magnetically. We give a detailed description of the low-temperature crystal structure. Polarized-neutron experiments show that the site susceptibilities of the two independent uranium atoms in U_2Pt_2In are substantially different and the conduction-electron polarization is unusually large. [S0163-1829(99)00118-6]

I. INTRODUCTION

The unusual properties exhibited by actinide intermetallic compounds, from spin fluctuators to superconducting heavy Fermions, have led to searches for new groups of compounds on which systematic measurements may be made. One such group has the generic formula An_2T_2X, where An is an actinide (U, Np, Pu, or Am), T is a transition metal (Co, Rh, Ni, Pd, Pt, etc.), and $X=Sn$ or In. First reports of the crystal structure and the stability of the phases was made by Mirambet *et al.*[1] and Peron *et al.*[2] in 1993. A more comprehensive review, including transuranium compounds, was given by Wastin *et al.* in 1995.[3] These systems were reported to crystallize in the tetragonal U_3Si_2 structure, which is illustrated in Fig. 1. The U-U distances in these materials approach the value of ∼3.5 Å, which is known as the Hill criterion[4] for direct $5f$-$5f$ hybridization effects to occur, so that a number of interesting electronic properties may be anticipated.

Shortly after their structural characterization, magnetic property measurements were reported on many of the compounds confirming a variety of different behavior.[5–13] In the last three years we have succeeded in growing single crystals of these materials in Karlsruhe; allowing a more detailed examination of the properties. In this paper we confine our attention to the three compounds with $T=Ni$, Pd, and Pt, and $X=In$. There are two major motivations for the present work. First, in the case of T=Ni and Pd the antiferromagnetic structure reported[14–16] from experiments on polycrystalline samples is rather complicated, being noncollinear in both cases. Since this is an unusual situation in actinide compounds (it is more common to find multi-**k** structures),[17] the exact magnetic structure is worth verifying with a single crystal. In previous studies with polycrystalline samples at most only four magnetic reflections were observed. In the case of the Ni compound, the authors[14] used this sparse information to not only refine the direction and magnitude of the uranium moment, but they further claimed a rather large ($0.37\mu_B$) moment on the nickel. Since a theoretical study of these materials[18] concluded that no moment should exist on the T sites, the presence of a Ni moment warrants further justification. Second, the Pt compound, which apparently does not order magnetically, has a large value for the electronic specific heat coefficient of 415 mJ/(mol U)K^2 which classifies this compound as a heavy Fermion uranium compound.[8,10] Moreover, recent measurements of the electrical resistivity and specific heat suggest that the compound displays non-Fermi liquid behavior at low temperature.[19–21] Although our measurements do not directly address this interesting point, we characterized the structure of the Pt compound at low temperature and show that no magnetic ordering occurs above 1.5 K. A polarized-neutron examination of the magnetization in the unit cell shows that the susceptibilities at the two independent U sites are different and that there is an unusually large conduction-electron polarization in the unit cell.

II. EXPERIMENTAL DETAILS

The crystals were grown at EITU, Karlsruhe, by a modified mineralization process using about 20 g of the molten sample encapsulated in a tungsten crucible and sealed under vacuum by electron-beam welding. All crystals were characterized by a four-circle x-ray diffractometer before being taken to Grenoble for the neutron experiments. The crystals were of irregular shape, with a weight of ∼20 mg. Most crystals were oriented with an **a** or **b** axis approximately vertical to allow the most independent reflections to be gathered. In the case of the Pt compound we examined two crystals from different batches to be sure that the parameters were independent of sample batch. The calculated linear ab-

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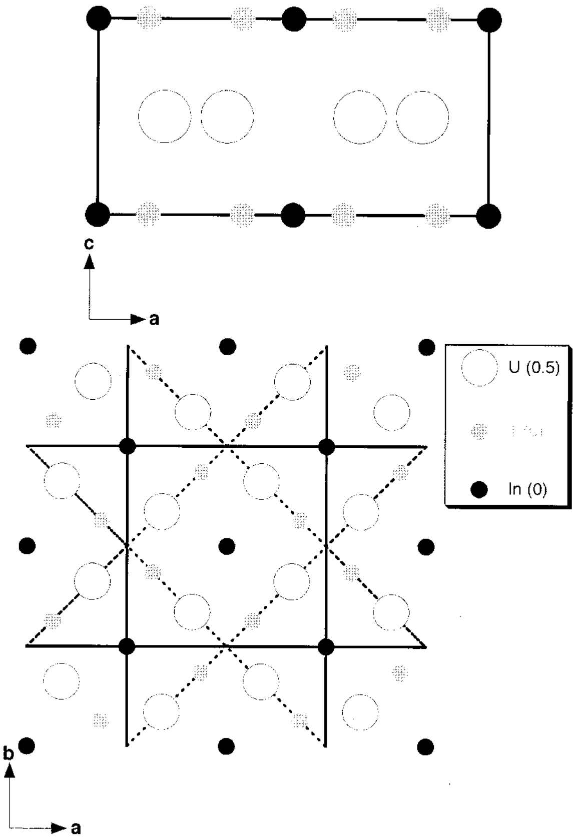

FIG. 1. Crystallographic structure for U_2T_2In (T=Ni and Pd), space group $P4/mbm$ (No. 127), in two different projections.

sorption coefficient was 1.20 cm^{-1}, arising almost totally from the In absorption, resulting in only small corrections.

The first set of measurements with $H=0$ were performed with the D15 normal-beam diffractometer with a lifting detector to allow access to much of reciprocal space. This diffractometer is installed at the Institut Laue Langevin's High Flux Reactor and uses a wavelength of 1.174 Å. Temperatures between 1.5 and 300 K were attained with a standard ''orange'' cryostat. An additional experiment was performed in a magnetic field ($H_{max}=6$ T) on the S-20 diffractometer (which has now been dismantled) to search for changes in the intensities of the magnetic reflections of T_N in the presence of a magnetic field.

In the case of the Ni compound the magnetic wave vector was independent of temperature and commensurate at $\mathbf{k} = (0, 0, 1/2)$, in agreement with previous work.[14–16] Intensities therefore could be collected easily without concern about the nuclear structure as the two contributions do not overlap. A total of 83 reflections, reducing to 21 inequivalent reflections were measured. In the case of the Pd compound $\mathbf{k} = (000)$, i.e., the magnetic and crystallographic cells are the same, again in agreement with previous work.[15,16] In this case we used the previously determined magnetic structure to calculate where the strongest peaks would be, and then collected the subset of magnetic reflections which did not fall on a strong nuclear reflection. To determine accurately the magnetic contribution, we measured reflections at 5 and 50 K, and by subtraction obtained 61 nonzero reflections, which reduced to a reliable set of 18 inequivalent magnetic intensities for the refinements.

The polarized-neutron measurements on U_2Pt_2In were made with the D3 diffractometer at the ILL on the same crystals used in the D15 experiments. A wavelength of 0.843 Å was used with an Er filter to reduce the $\lambda/2$ contamination. The crystal was at $T=10$ K and a magnetic field of 4.6 T applied in the [101] direction. Standard ''flipping ratios'' were measured and these allow a determination of the ratio of the (small) magnetic contribution to the nuclear reflections. Since the D15 experiments establish the nuclear structure, the magnetic contributions can be deduced and thus the magnetization within the unit cell. The *total* (bulk) magnetization at this H and T is $0.184\mu_B$/per mole.[22]

III. RESULTS

A. Crystallography

We have already shown the U_3Si_2 structure of the space group $P4/mbm$ ($Z=2$) in Fig. 1. As expected, our structural refinements (Table I) for T=Ni and Pd are in excellent agreement with those obtained from a Rietveld analysis of the polycrystalline compounds.[14–16] Interestingly, the atomic positions we obtain are closer to those reported from the pulsed neutron study[14,15] (in fact our values are *exactly* the same to four significant figures) suggesting that the superior Q range normally associated with this technique gives a better value for the structural parameters. The extinction in all of these crystals is reasonably small, with $y_{min}\sim 0.74$ in the case of the Ni compound.

In the case of the Pt compound the structure is not the simple $P4/mbm$. Instead, as found first by x rays working on a small crystal of this material,[22] it crystallizes in the space group $P4_2/mnm$ ($Z=4$), which corresponds to doubling the unit cell in the **c** direction and shifting slightly the atoms from their symmetric positions as shown in Fig. 2. This structure, the so-called Zr_3Al_2 type, was first reported for U_2Pt_2Sn,[23] and may be regarded as a superstructure of the U_3Si_2 structure. As seen in Fig. 2 (and noted in Table I) there are now two inequivalent U sites, those at sites $4g$ (U_1) and $4f$ (U_2) and in Table II we give the coordination sphere around each of these uranium atoms. Since the U atoms remain on the planes at $z=0$ and 0.5, it is clear from Fig. 2 that the one single distance of the U atoms that exists in the high symmetry $P4/mbm$ structure (see Fig. 1) now becomes *two* inequivalent distances. As we shall discuss later these distances may play an important part in defining the magnetism of these materials.

Our refinements show that all structures are stoichiometric in the sense that no significant improvements in the R factors could be achieved by allowing the occupational parameters to vary. This is important particularly with respect to the Pt compound and the appearance of the non-Fermi liquid state at low temperature.[19–21] Many materials that exhibit this phenomenon are actually disordered compounds, such as $CeCu_{5.9}Au_{0.1}$,[24] and there has been some debate whether the two phenomena are related.[25] At least in U_2Pt_2In this matter is not an issue.

The reduced space group of the Pt compounds gives systematic absences at the positions ($h0l$) with $h+l$ odd. An examination of the data collection shows that these reflec-

TABLE I. Results of the crystal structure refinements. Two crystals from different batches were examined for U_2Pt_2In. $\langle y \rangle$ is the mean value of extinction refined, and y_{min} corresponds to the largest reduction (compared to an unaffected intensity with $y=1$) calculated from extinction. The In atoms are fixed by symmetry to the positions $2a$ in the $T=$Ni and Pd compounds, and $4d$ for $T=$Pt. Values of the coherent scattering lengths used (in Fermis) are U=8.417, In=4.065, Ni=10.3, Pd=5.91, Pt=9.60.

	U_2Ni_2In ($T=25$ K)	U_2Pd_2In ($T=50$ K)	U_2Pt_2In ($T=20$ K)	U_2Pt_2In ($T=75$ K)
Weight (u.m.a.)	708.3	803.68	981.06	981.06
Space group	$P4/mbm$	$P4/mbm$	$P4_2/mnm$	$P4_2/mnm$
a (Å)	7.390(5)	7.623(5)	7.684(5)	7.708(5)
c (Å)	3.587(5)	3.739(5)	7.335(5)	7.371(5)
V (Å^3)	195.867	217.274	433.065	437.951
Z	2	2	4	4
x_U ($4h$)	0.1728(1)	0.1745(1)		
x_T ($4g$)	0.3742(1)	0.3713(1)		
x_{U_1} ($4g$)			0.1832(1)	0.1829(2)
x_{U_2} ($4f$)			0.3397(1)	0.3387(2)
x_T ($8j$)			0.1306(1)	0.1305(2)
z_T ($8j$)			0.2273(1)	0.2286(2)
Reflections				
measured	262	192	693	215
Unique reflections				
with $I>3\sigma(I)$	88	74	301	87
Θ range	6 – 66	6 – 40	2 – 60	4 – 45
Extinction				
g (10^{-4} rad^{-1})	0.049(4)	0.042(6)	0.058(4)	0.014(5)
$\langle y \rangle$	0.96	0.98	0.99	0.99
y_{min}	0.74	0.86	0.79	0.94
Refinement				
R	0.031	0.024	0.026	0.036
χ^2	3.4	0.9	2.2	2.2
No. of variables	7	7	10	10

tions always had a small (about 0.1% of the strongest reflection) intensity. However, given the very few systematic absences in this structure, there is a strong possibility for these reflections to be affected by multiple scattering, especially since the wavelength on D15 is quite short (1.174 Å). Since intensities of all these reflections were similar, and greater than arising from any second-order contamination (of $\sim 3 \times 10^{-4}$), we have neglected them in the refinement, and assumed they originate from multiple scattering. A subsequent test experiment on the D10 diffractometer at the ILL which has the capability of allowing a sample rotation about the scattering vector while examining the diffracted intensity showed that the intensities were very dependent on the rotation angle ψ. An examination of some 20 of these weak reflections allows us to conclude that the intensity observed on D15 was indeed caused by multiple scattering, and neglecting these reflections in the analysis is justified at the level of 1 part in 10^4 of the strong reflections.

The staggering of the uranium chains (lower part of Fig. 2) actually *decreases* one of the U-U distances, and, of course, increases the other. These distances, within the basal plane of the tetragonal structure are known as $d_{\parallel}$, whereas the distances along the tetragonal **c** axis are known as $d_{\perp}$ (see Fig. 1 of Ref. 15). For the $T=$Ni and Pd compounds the values of $d_{\parallel}$ and $d_{\perp}$ are straightforward and are given in Table III. In the Pt compound it is less simple. The distortion of the rows (parallel) to the tetragonal **c** axis, allows us to define a mean position $\overline{x_U}$, which, if occupied by both U atoms, would result in a single column of U atoms parallel to the tetragonal axis. $\overline{x_U}$ is defined such that the U atoms are displaced an equal distance Δx_U on each side of it. The same arguments may be made for the Pt z coordinates, similarly defining a displacement of Δz_T. All values are given in Table III. As a function of temperature, the Δx_U and Δz_T values show that the overall distortion of the structure actually increases as the temperature is lowered. Usually, atomic positional parameters are stable as a function of temperature, and it is the lattice parameters that change; the present findings probably indicate considerable anharmonicity. At some higher (than room) temperature the U_2Pt_2In structure may transform into the higher symmetry $P4/mbm$ space group.

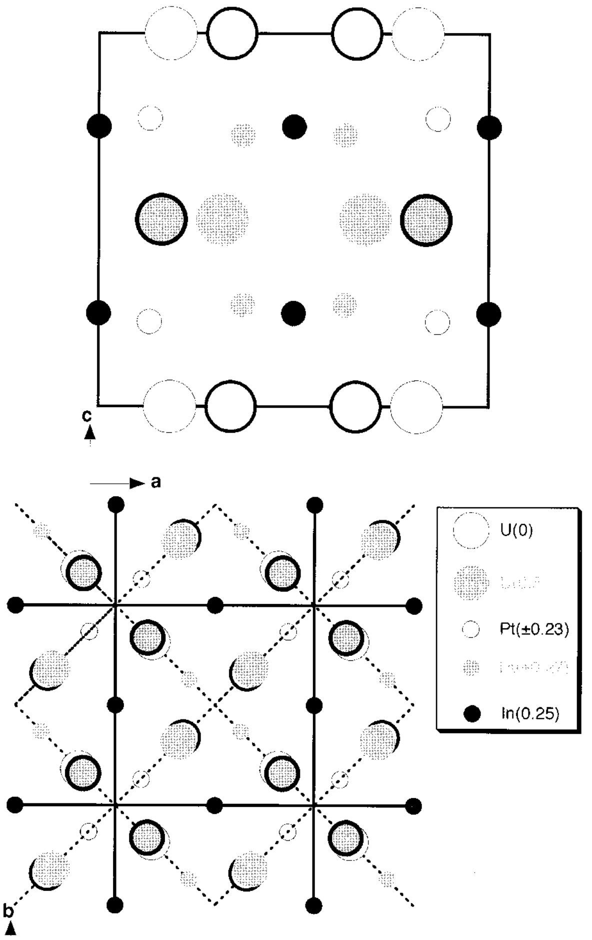

FIG. 2. Crystallographic structure of U_2Pt_2In compound, space group $P4_2/mnm$ (No. 136), in two different projections. The U_2 atoms at the $4f$ site are marked with bold borders; these are the atoms that have a separation of only 3.48 Å in this structure.

B. Magnetic structure

As first derived by Bouree *et al.*,[26] Fig. 3 shows the possible magnetic structures in the $P4/mbm$ space group with the magnetic propagation vector $\mathbf{k}=(000)$. The χ^2 values obtained by refining the data with the different structures for the $T=$Ni and Pd samples are given in Table IV. For the form factors we have used the U^{3+} configuration and the dipole approximation from the tables in Freeman *et al.*[27]

The χ^2 values in Table V show large differences and leave no doubt that the correct structures are Γ_8 for $T=$Pd and Γ_3 (equivalent to Γ_2 in Fig. 3) for $T=$Ni. The single crystal data are, as expected, far more sensitive than the refinements on polycrystalline materials, but it is important to stress that, despite the difficulty of only a few magnetic reflections, the early attempts did indeed obtain the *correct* magnetic structures. On the other hand, the values of the uranium moments proposed previously are systematically low. For $T=$Ni, the values were 0.60(1) (Ref. 14) and $0.85(5)\mu_B$ (Ref. 16), whereas our value is $0.92(2)\mu_B$. For $T=$Pd, the values were 1.6(2) (Ref. 15) and $1.55(5)\mu_B$ (Ref. 16), whereas our value is $1.73(1)\mu_B$. Our refinements show *no* evidence for any moments on the T sites with a precision of about $0.04\mu_B$. Thus the observation of a moment of $\sim 0.4\mu_B$ at the Ni site in Ref. 14 is probably a consequence of unjustified confidence in the result of the Rietveld refinement.

TABLE II. Interatomic distances around the U_1 and U_2 atoms in U_2Pt_2In calulated at 20 K. NN refers to the number of neighbors. The standard deviations are ± 0.005 Å.

Atom	NN	Atom	d (Å)	Atom	NN	Atom	d (Å)
$U_1(4g)$	2	$U_2(4f)$	3.676	$U_2(4f)$	1	$U_2(4f)$	3.484
$U_1(4g)$	2	$U_2(4f)$	3.858	$U_2(4f)$	2	$U_1(4g)$	3.676
$U_1(4g)$	2	$U_2(4f)$	4.194	$U_2(4f)$	2	$U_1(4g)$	3.858
$U_1(4g)$	1	$U_1(4g)$	3.982	$U_2(4f)$	2	$U_1(4g)$	4.194
$U_1(4g)$	2	Pt$(8j)$	2.845	$U_2(4f)$	2	Pt$(8j)$	2.819
$U_1(4g)$	4	Pt$(8j)$	2.960	$U_2(4f)$	4	Pt$(8j)$	3.008
$U_1(4g)$	4	In$(4d)$	3.357	$U_2(4f)$	4	In$(4d)$	3.420

For single crystals of $T=$Pt exhaustive scans along different reciprocal lattice lines at 1.5 K, revealed no evidence of any long- or short-range magnetic ordering. These findings are in agreement with bulk experiments, in which no evidence for a cooperative phase transition has been found.[8–11] Recently muon experiments[21] also exclude any small *ordered* moment, although, interestingly, they do suggest the presence of antiferromagnetic *fluctuations*.

C. Temperature dependence of moments

The temperature dependence of the square of the magnetic moment is shown in Fig. 4 and Fig. 5 for $T=$Ni and $T=$Pd, respectively. The values of T_N deduced are in good agreement with those reported from bulk measurements in Refs. 14 and 15, 14 and 43 K, respectively, but are higher than those found by neutron diffraction and reported in Ref. 16 for the $T=$Pd compound, in which $T_N \sim 31$ K. However, very few points were taken as a function of temperature.

The exponent of the magnetization below T_N are 0.28 (1) and 0.34 (2) for the Ni and Pd compound, respectively. These values are consistent with most three-dimensional systems, and are commonly found in anisotropic uranium compounds. For comparison, a value of $\beta=0.28$ (3) was found for the isostructural compound U_2Rh_2Sn.[28] The magnetic structure in this material is Γ_5 of Fig. 3, i.e., the moments are along the **c** axis.

High-field experiments on U_2Pd_2In (Ref. 29) have shown that at magnetic fields of between 23 T (at $\sim$20 K) and 27 T there is a metamagnetic phase transition; i.e., the ferromagnetic component suddenly increases. Such fields are not available together with neutrons. In an attempt to see whether the magnetic structure is unstable with lower fields but at temperatures near T_N, we have tried cooling the material through T_N in a magnetic field of 6 T applied along the **a** axis, but no changes were observed.

D. Polarized-neutron experiment on U_2Pt_2In

As discussed above, the U_2Pt_2In structure has two inequivalent sites, $U_1(4g)$ and $U_2(4f)$ with slightly different coordination spheres, see Table II and Fig. 2. The closest U atom to U_2 is a similar U_2 atom at $d_{\parallel}=3.484$ Å at 20 K. Note that the interatomic distances given in Ref. 22 are at 300 K;

TABLE III. The distortions in the U_2Pt_2In as compared to T=Ni and Pd. $\overline{x_U}$ is the position which would correspond to all U atoms being on rows parallel to the tetragonal **c** axis. For the Ni and Pd compounds, this is the real value of x_U (see Table I), whereas for the Pt compound it is taken so that the atoms are displaced a constant amount Δx_U either side. The same arguments can be made for the Pt atoms which are displaced in the **c** direction. $d_{\parallel}$ is the distance in the basal plane between nearest U neighbors, $d_{\perp}$ is the distance (to a good approximation) along the **c** axis. With the inequivalence of the U_1 and U_2 in the Pt structure there are two values of $d_{\parallel}$. The shortest bonds are marked in bold.

	$d_{\parallel}$ (Å)		$d_{\perp}$ (Å)	$\overline{x_U}$	Δx_U	Δz_T
U_2Ni_2In	3.612(5)		**3.587(5)**	0.1728(1)		
U_2Pd_2In	3.762(5)		**3.739(5)**	0.1745(1)		
U_2Pt_2In (20 K)	**3.484(5)**	3.982(5)	3.676(5)	0.1718(2)	0.0115(2)	0.0227(2)
U_2Pt_2In (75 K)	**3.571(5)**	3.987(5)	3.686(5)	0.1721(4)	0.0108(4)	0.0214(2)
U_2Pt_2In (300 K)	**3.583(1)**	3.925(1)	3.6878(3)	0.1725(1)	0.0079(1)	0.0155(1)

because of the further distortion of the structure as the temperature is lowered, the parameters at low temperature, where the interesting physics occurs, are slightly different. The closest U neighbors for the U_1 site are *two* U_2 neighbors in the [001] direction $d_{\perp}=3.676$ Å. Our polarized-neutron measurements probe the individual site susceptibility of these two U atoms, and the results are given in Table V. The observed magnetization density in the unit cell, constructed using the maximum entropy method,[30] is shown in Fig. 6.

The most important, and surprising, result is that the site susceptibility of U_2 is almost double that of U_1. Since the ‘‘average’’ U-U distances (obtained by averaging over the nearest 7 U neighbors of Table II) are not too different for the two atoms (3.85 Å for U_2 and 3.92 Å for U_1) the factor of two in the site susceptibility is surprising. As shown in Ref. 22 the bulk susceptibility is not strongly anisotropic (a factor of ~15% at low temperature), so that the significant difference in the site susceptibility probably arises from details of the U-U and U-Pt hybridization processes.

In the analysis we also deduce the amount of $\langle j_2 \rangle$ contribution to the form factor, and this allows an estimate of the ratio of the orbital and spin magnetic moments.[31] These values (Table V) are the same for the two sites, and correspond to values frequently found for U ions in intermetallic compounds,[32] and reported, for example, for URhAl.[33] For the free-ion U^{3+} configuration the ratio $|\mu_L/\mu_S|=2.57$, so that a value of ~1.8 represents an appreciable quenching of the orbital moment, as is expected when hybridization occurs.[31]

No significant susceptibility is found at the Pt site. This is not surprising; even if the Pt $5d$ states were polarized (see below) they would have a form factor that falls rapidly with scattering angle, and would be difficult to detect in our experiment. The individual site susceptibilities may be added and compared to the bulk susceptibility as given in Ref. 22. The neutron value of the total induced moment (Table V) is significantly lower than that found by the bulk measurements (by some 25%). In compounds such as US,[34] the moment observed with neutrons is larger (by ~10%) than that observed with magnetization. This is because the bulk measurements observe all the magnetization in the unit cell, i.e., that associated with the localized $5f$ electrons, as well as that arising from the conduction-electron states, and for magnetic elements with less than a half-filled electron shell, these are aligned antiparallel. The neutron measurement is sensitive to only that part of the magnetization associated with the localized $5f$ states, so that it is normally *greater* than that obtained by bulk methods; the difference attributed to the negative conduction-electron polarization.[34] Thus, the large positive discrepancy in the case of U_2Pt_2In is unusual. At least two possibilities exist for this additional conduction-electron polarization; it can arise from either the Pt $5d$ elec-

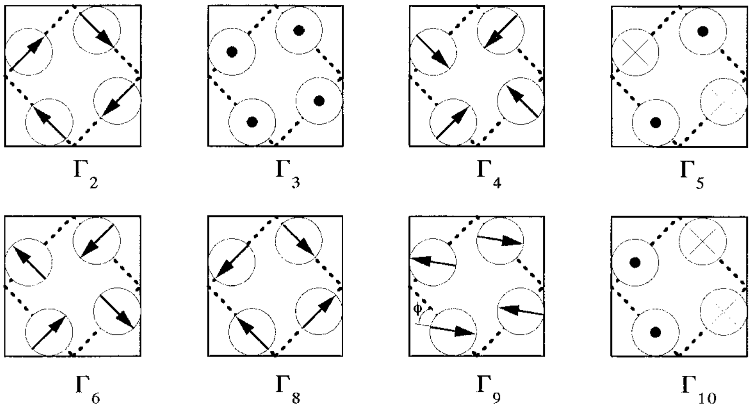

FIG. 3. Possible magnetic arrangements in the space group $P4/mbm$ with the representations corresponding to $\mathbf{k}=(000)$ — taken from Ref. 26. For $\mathbf{k}=(0, 0, 1/2)$, the arrangements are the same, but their representation changes. The correspondence between the two is given in Table IV.

TABLE IV. For convenient reference to Fig. 3 the representations are in the order equivalent to **k** $=(000)$, which corresponds to U_2Pd_2In, whereas U_2Ni_2In has **k** $= (0, 0, 1/2)$. For identical arrangements of one layer of U moments (as shown in Fig. 3) the representations are shown in each column. Given are the goodness-of-fit χ^2 values for the refinements and the resulting uranium moment. In the final column for each case are the results of a refinement including the best configuration (lowest χ^2) of the U moments and allowing a moment on the T site. The symmetry of the T site moment is given by the second representation in the column heading. For $T=$Ni, 83 reflections were measured, reducing to 19 inequivalent structure factors with $I>3\sigma(\mathrm{I})$. For $T=$Pd, the corresponding numbers were 61 and 18. Note that with the possibility of domains in the tetragonal basal plane, it is not possible to distinguish any value of ϕ in the arrangement labeled Γ_9 in Fig. 3.

U_2Pd_2In	Γ_2	Γ_3	Γ_4	Γ_5	Γ_6	Γ_8	Γ_9	Γ_{10}	Γ_8/Γ_{10}
χ^2	80.5	472	37.8	299	165	**1.40**	234	131	1.35
U moment (μ_B)	1.67(8)	1.4(2)	1.75(6)	2.0(2)	1.6(1)	**1.73(1)**	1.5(1)	2.1(1)	1.74(1)
Pd moment (μ_B)									0.025(15)
U_2Ni_2In	Γ_3	Γ_2	Γ_1	Γ_8	Γ_7	Γ_5	Γ_{10}	Γ_9	Γ_3/Γ_2
χ^2	**4.18**	1770	213	970	109	192	728	421	4.14
U moment (μ_B)	**0.918(4)**	0.7(1)	1.03(3)	1.4(1)	0.95(2)	0.97(3)	1.14(8)	1.12(6)	0.919(4)
Ni moment (μ_B)									0.029(24)

trons or a fraction of the U $5f$ states that are delocalized. As far as we are aware in uranium compounds, it is only in UPd_2Al_3 — a well-known heavy-Fermion superconductor — where a positive conduction-electron polarization has been found.[35]

IV. DISCUSSION

Our study confirms the magnetic structures of previous work on polycrystalline samples, but with important differences. In particular, our values of the magnetic moments are larger (by $\sim 15\%$) than those published earlier. Since the structures of the Ni and Pd compounds are *noncollinear* this confirmation is important, especially when the previous refinements were performed on only four magnetic reflections. Such noncollinear magnetic structures are unusual in the actinides. Sandratskii and Kübler[36] have discussed this problem in general, and in the particular case of U_2Pd_2Sn have shown that the strong spin-orbit coupling present in the actinides stabilizes the noncollinear magnetic arrangement. An important constraint is the point symmetry at the position of the U atoms; once the moments point in the tetragonal basal plane, the most likely magnetic structures are those compatible with the symmetry, and these are noncollinear.

Some success has been achieved in understanding the anisotropy in actinide compounds as arising from a hybridization-mediated mechanism that depends on the nearest approach of An-An neighbors. Thus, if the actinide atoms are stacked in a tightly packed basal plane arrangement, as is the case, for example, in the 1:1:1 UTX compounds,[37,38] then strong hybridization is set up in the basal plane and it is energetically favorable to find the moment *perpendicular* to this hybridization-mediated bonding. In the present compounds the bond lengths are not very different $d_\perp$ (the shortest U-U distance perpendicular to the plane) being 3.59 Å for $T=$Ni, and 3.74 Å for $T=$Pd, see Table III. The correspond-

TABLE V. Refinements for the polarized-neutron values for U_2Pt_2In. The experiment was performed at $T=10$ K with an applied field of 4.6 T. 161 reflections were measured with $\lambda=0.843$ Å. The data have been analyzed within the dipolar approximation with the single-electron wave functions of Freeman *et al.* (Ref. 27). The induced magnetic moment from the magnetization measurements (on crystals from the same batch) for these conditions is $0.184(4)\mu_B$ per formula unit. μ_L and μ_S are the orbital and spin moments, respectively. The value for μ_L/μ_S for the U^{3+} free ion is -2.57.

	U_1 $(4g)$	U_2 $(4f)$
Moment $\mu(m\mu_B)$	47 (9)	92 (9)
Orbital moment $\mu_L(m\mu_B)$	100 (20)	220 (20)
$-\mu_L/\mu_S$	1.8 (5)	1.7 (2)
Total moment $(m\mu_B)$	140 (10)	
Bulk moment $(m\mu_B)$	184 (4)	
Conduction-electron polarization $(m\mu_B)$	+ 44 (12)	

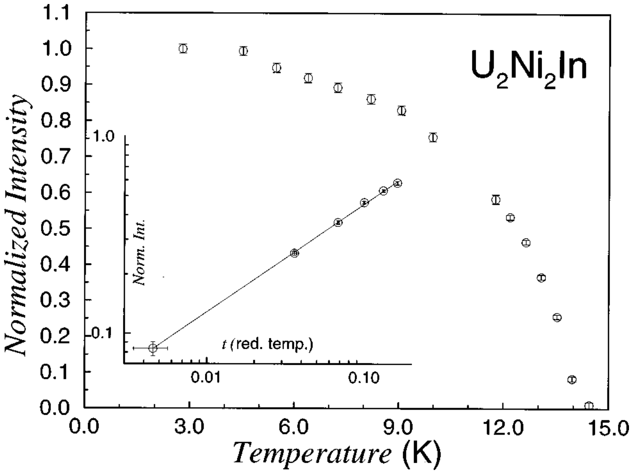

FIG. 4. Temperature dependence of the intensity of the (1, 0, 1/2) magnetic reflection in U_2Ni_2In shown on a scale normalized to the value at 2.5 K. The inset is a log-log plot which gives $T_N=14.0$ (1) K, and the exponent $\beta=0.28$ (1). The reduced temperature $t=(T-T_N)/T_N$.

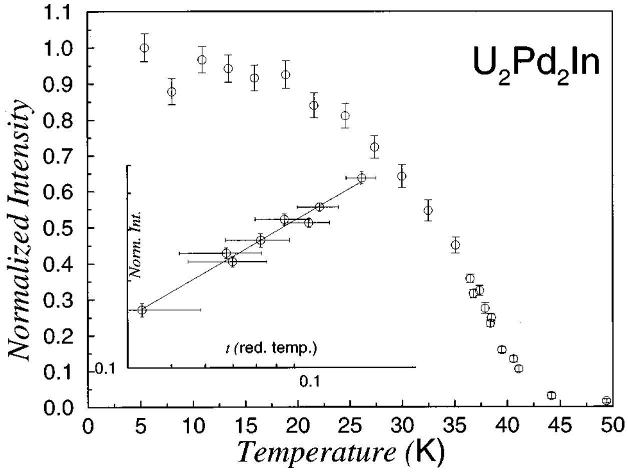

FIG. 5. Temperature dependence of the intensity of the (1, 0, 1) magnetic reflection in U_2Pd_2In shown on a scale normalized to the value at 4 K. The inset is a log-log plot which gives $T_N = 40.8$ (7) K, and the exponent $\beta = 0.34(2)$.

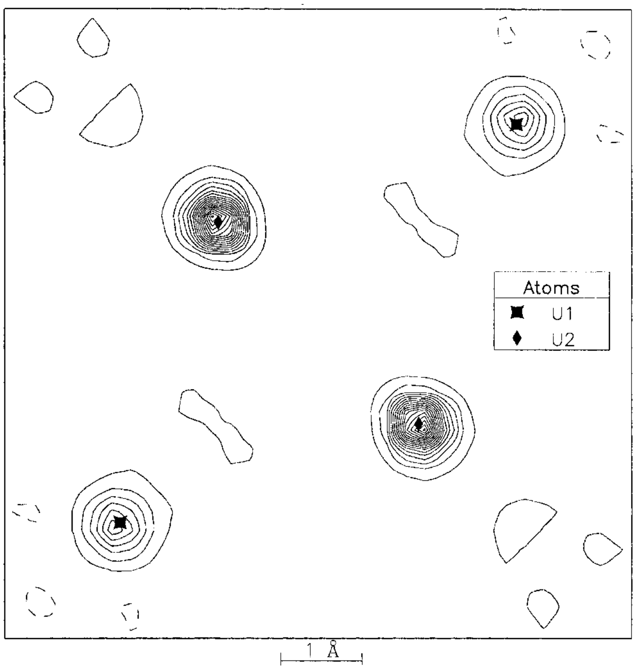

FIG. 6. (001) projection showing the magnetization density (determined by the maximum entropy method) induced at $T = 10$ K with $H = 4.6$ T in U_2Pt_2In. Note the larger signal at the U_2 $(4f)$ compared to that at the U_1 $(4g)$ site. Contour levels are from -0.01 (dashed) to 0.41 in steps of $0.02\mu_B/\text{Å}^3$.

ing $d_\parallel$ (within the basal plane) distances are 3.61 and 3.76 Å, respectively, so that it seems unlikely that such a simple model (which does not even take into account the number of neighbors) is valid with such small differences. Perhaps by chance, the moments in these compounds do in fact lie perpendicular to the shortest U-U distance, although this is not the case with the compound U_2Rh_2Sn,[28] in which the moments point along the **c** axis, even though $d_\perp = 3.63$ Å is the closest U-U distance. However, one should be cautious in taking these ideas too literally; probably more important is the filling of the d band and the consequent hybridization with the $5f$ states. The Rh-Sn compound is the only one in this isoelectronic column that orders magnetically. This may well be the most important parameter as those with Ni and Pd, which are isoelectronic, with either In or Sn appear to order with the moments in the basal plane (the case of the Pt-Sn compound is not yet resolved) giving possibly the first hint of systematics involving the T element and the occupation of its d band.

The previous work on U_2Ni_2In (Ref. 14) obtained a moment of $\sim 0.4\mu_B$ on the Ni site. Our refinements with single crystals show that the moment is $0.03(2)\mu_B$, i.e., zero. This is in agreement with arguments by Divis *et al.*[18] in which they predict no moments on the T atoms in these compounds.

U_2Pt_2In has a modified crystal structure as compared to the $T = $ Ni and Pd compounds, but the refinements show no evidence for lack of stoichiometry or atomic disorder. We have discussed this structure in detail as the compound exhibits non-Fermi-liquid behavior at low temperature.[19–21] It does not order magnetically. We have performed a polarized-neutron experiment with the sample at $T = 10$ K, and an applied field of 4.6 T to measure the induced magnetization in the unit cell. There is a substantially different site susceptibility for the two independent U sites, with U_2 site (see Tables I and II, and Fig. 2) having the larger value. Although this crystal structure is rather complicated, it will be interesting to see whether these unusual features can be reproduced by theory. Moreover, the large conduction-electron density deduced from comparing the results of the magnetization and polarized-neutron experiments may be relevant to the unusual transport properties of this material at low temperature.

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