

Magnetism of UT_2Si_2 ($T=Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Os$) from spin-density-functional calculations

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(Received 7 April 1994)

The electronic structure of UT_2Si_2 , where $T=Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Os$, is determined by means of self-consistent density-functional calculations in the local approximation treating the U 5*f* states as band states. A pronounced trend in the hybridization strength is exposed and is deduced to give rise to the different magnetic properties of the compounds. In agreement with experiment, the nonmonotonic behavior in the subseries containing 3*d* elements is reproduced, especially the state of UFe_2Si_2 which is nonmagnetic in contrast to the other 3*d* members. The underlying physics of this fact is explained. Furthermore, special features of the heavy-fermion system URu_2Si_2 are discussed.

I. INTRODUCTION

Ternary intermetallic compounds UT_2Si_2 (T denotes a transition metal) show a rich variety of electronic and magnetic ground state properties, including Pauli paramagnetism and ferro- and antiferromagnetism. In almost all cases only the U atom is observed to be magnetic, although in the case of $T=Mn$ (and probably also Cr) the 3*d* atoms also carry a nonvanishing magnetic moment.

In spite of the considerable interest that these compounds received (see, e.g., Refs. 1–3), it is still not clear which physical mechanism determines their magnetic properties. The question is whether or not their magnetic behavior can be successfully described within the local spin-density-functional (LSDF) (Refs. 4, 5) approach or whether it is governed by a Kondo-type screening of the uranium local moments via the valence electrons of the ligands.

In a recent paper⁶ Endstra *et al.* concluded that, because of the nonmonotonic dependence of the temperature of the magnetic phase transition on the strength of the hybridization between the U 5*f* states and the ligand *d* states, a Kondo-type interaction is responsible for the ground state properties of these compounds. However, this nonmonotonic behavior cannot be taken as a decisive criterion because a similar behavior can be obtained within the LSDF theory. Indeed, an increase of the hybridization in a system with well-defined U 5*f* moments leads to a stronger magnetic interaction between the moments through a Ruderman-Kittel-Kasuya-Yosida- (RKKY-) type mechanism and, therefore, to a higher transition temperature. However, a further increase of the hybridization can make the local moment of the U atom ill defined and, finally, a nonmagnetic state of the crystal becomes energetically preferable.

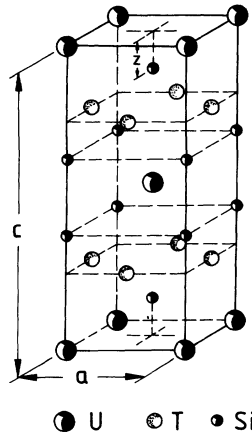
In the present paper, a rather broad overview where a multitude of details are omitted, we report results of LSDF calculations for the ten compounds UT_2Si_2 where $T=Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Os$. We believe that

this study is of special interest because of the differences in the magnetic states of these compounds at low temperatures and because of the heavy-fermion and superconducting properties of URu_2Si_2 . Although the simplified way of our treatment of many-body effects by the LSDF theory precludes a proper description of heavy-fermion behavior and superconductivity, the study of the difference in the band structure of these compounds and comparison of theoretical with experimental features supply important information about the physical properties of the compounds providing a useful basis for a more refined treatment of many-body effects. Furthermore, material specific properties can only be obtained by first principles electronic structure calculations of the kind we present here.

II. CALCULATIONAL RESULTS AND DISCUSSION

All the compounds under investigation here crystallize in the body-centered-tetragonal $ThCr_2Si_2$ structure shown in Fig. 1. Our calculations were carried out with the augmented spherical wave (ASW) method⁷ including the effects of spin polarization and spin-orbit interaction treating all valence electrons on an equal basis, i.e., in particular, treating the uranium 5*f* electrons as band electrons. As far as the magnetic order is concerned our calculations were done for assumed nonmagnetic, ferromagnetic, and antiferromagnetic compounds where the moment was chosen parallel to the *c* axis. For the lattice parameters the experimental values were used; they are collected in Table I together with the relevant references. To facilitate comparison the radii of the atomic spheres of U and Si throughout were chosen to be 1.90 Å and 1.39 Å, respectively, for all compounds. For the transition metal T , the radii are given in Table I.

We begin with Table II which contains experimental and theoretical information about the presence or absence of magnetic moments on the U and T atoms in the

FIG. 1. Unit cell of the $ThCr_2Si_2$ crystal structure.

ground state of the series of compounds UT_2Si_2 . Of special interest is the magnetic behavior within the subseries of compounds where the T atom is a $3d$ transition element, i.e., the first six compounds in Table II. Indeed, here we observe “nonmonotonic” magnetic properties. For the Cr and Mn compounds not only the U atom but also the $3d$ atom have the tendency to be magnetic. For the next member of the series, the Fe compound, both atoms are nonmagnetic. The remaining three compounds possess a magnetic U atom. The calculated values of the atomic spin and orbital moments in the magnetic compounds are collected in Table III. They were obtained for the experimentally observed magnetic order, that is, ferromagnetic for (UMn_2Si_2 and UCu_2Si_2) and antiferromagnetic for (UCr_2Si_2 , UCo_2Si_2 , UNi_2Si_2 , URh_2Si_2 , UPd_2Si_2).

Returning to Table II we note the good agreement between the experimental and theoretical results. The only inconsistency is obtained for URu_2Si_2 where our calculations fail to reproduce the small experimental magnetic moment of $0.04\mu_B$ (Ref. 1) giving a nonmagnetic state. The compound URu_2Si_2 is unusual as it is the only compound in this series exhibiting both heavy-fermion and superconducting properties. The simplified way of the

TABLE I. Parameters used for the calculations. Here a , c , and z are the crystal structure parameters; r_T is the radius of the atomic sphere of the transition metal T .

Ref.	a (Å)	$\frac{c}{a}$	z (c)	r_T (Å)	
UCr_2Si_2	3	3.91	2.69	0.38 ^a	1.52
UMn_2Si_2	8	3.92	2.62	0.39	1.50
UFe_2Si_2	8	3.95	2.41	0.37	1.41
UCo_2Si_2	9	3.92	2.45	0.37	1.39
UNi_2Si_2	9	3.96	2.40	0.37	1.41
UCu_2Si_2	9	3.99	2.50	0.38	1.50
URu_2Si_2	10	4.13	2.32	0.38 ^a	1.54
URh_2Si_2	11	4.01	2.50	0.38	1.52
UPd_2Si_2	12	4.08	2.46	0.38	1.57
UOs_2Si_2	13	4.12	2.33	0.38 ^a	1.54

^aExperimental value is not reported. A value in the range of those of the other compounds is used.

TABLE II. Magnetic state of the U and T atoms in UT_2Si_2 compounds. Experimental information is taken from Ref. 6. The theory entry represents the results of our calculations. Magnetic atoms are indicated with +, nonmagnetic atoms with -; question mark (?) denotes unclear experimental situation.

	U atom		T atom	
	Experiment	Theory	Experiment	Theory
UCr_2Si_2	+	+	?	+
UMn_2Si_2	+	+	+	+
UFe_2Si_2	-	-	-	-
UCo_2Si_2	+	+	-	-
UNi_2Si_2	+	+	-	-
UCu_2Si_2	+	+	-	-
URu_2Si_2	+	-	-	-
URh_2Si_2	+	+	-	-
UPd_2Si_2	+	+	-	-
UOs_2Si_2	-	-	-	-

treatment of quantum many-body effects by the LSDF theory precludes a proper description of heavy-fermion (HF) behavior and superconductivity. We assume that it is the same type of many-body interactions that is responsible for the small magnetic moment thus requiring a more refined treatment than we are able to give here.

To understand the origin of the differences in the magnetic properties we carried out calculations for the nonmagnetic states for all compounds. Our calculated elec-

TABLE III. Number of the U $5f$ electrons, n_{5f} , and T d electrons, n_d , and the values of spin m_s and orbital m_{or} atomic magnetic moments in UT_2Si_2 compounds. Only finite magnetic moments are given. For UCr_2Si_2 , UCo_2Si_2 , UNi_2Si_2 , URh_2Si_2 , UPd_2Si_2 magnetic structure is antiferromagnetic; for UMn_2Si_2 and UCu_2Si_2 it is ferromagnetic. The direction of the U spin moment is chosen as a positive direction.

	U atom		T atom	
	n_{5f}	m_s	n_d	m_{or}
UCr_2Si_2	2.84		4.71	
	1.08	-1.33	0.74	-0.46
UMn_2Si_2	2.84		5.73	
	1.47	-1.52	-1.90	-0.06
UFe_2Si_2	2.79		6.73	
UCo_2Si_2	2.85		7.68	
	1.17	-1.79	-	-
UNi_2Si_2	2.83		8.65	
	1.40	-2.15	-	-
UCu_2Si_2	2.89		9.44	
	2.21	-3.09	-	-
URu_2Si_2	2.87		6.70	
URh_2Si_2	2.88		7.67	
	1.35	-2.34	-	-
UPd_2Si_2	2.88		8.68	
	1.78	-2.57	-	-
UOs_2Si_2	2.93		6.32	

tron densities of states (DOS) are shown in Figs. 2–4.

The main differences in the DOS are seen to be the relative energy positions of the d states of the T atom compared with the uranium $5f$ states. Analysis of the $3d$ (Figs. 2 and 3) and $4d$ [Figs. 4(a),(b),(c)] series of compounds shows a clear tendency for the d bands to shift to lower energies relative to the $5f$ bands with increasing atomic number of the transition element. Indeed, for both UCu_2Si_2 and UPd_2Si_2 the T d states lie lower than the U $5f$ states and are separated from them by an energy interval of about 4 eV (0.3 Ry) and 2 eV (0.15 Ry), respectively. This interval becomes smaller with decreasing atomic number. In the case of URu_2Si_2 for the $4d$ series and starting with UFe_2Si_2 for the $3d$ series both groups of states form one common energy region and only the partial DOS allow us to distinguish the nature of states in the lower and upper parts of this region. This strong dependence of the relative energy position of the U $5f$ states and the T d states on the type of the T atom was observed also for another series of U compounds^{14,15} (see also the discussion in Ref. 16) and is caused by an increasing spatial localization of the d states in atoms with increasing atomic number. Note that the only $5d$ compound considered in the present paper, UOs_2Si_2 , shows no energy separation between d and f bands.

The smaller energy separation between the d and f states leads to stronger hybridization between these states. This hybridization changes substantially the structure of the $5f$ DOS and, as we believe, is responsible for the nonmagnetic ground state of UFe_2Si_2 , URu_2Si_2 , and UOs_2Si_2 obtained in our calculations.

In fact, in the case of UCu_2Si_2 , where the d - f hybridization is smallest, one can easily see that the $5f$ states are divided into two groups [Fig. 1(c)]. This split-

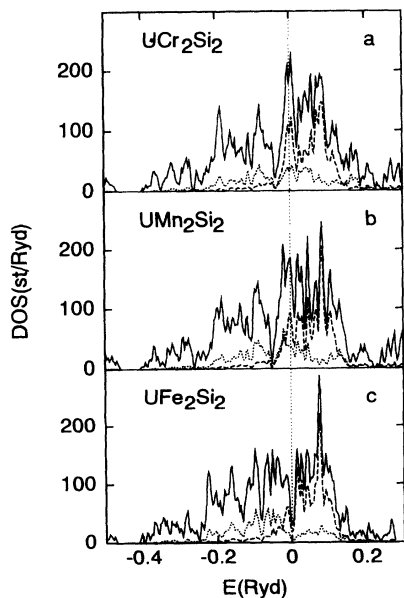


FIG. 2. DOS of nonmagnetic UCr_2Si_2 (a), UMn_2Si_2 (b), and UFe_2Si_2 (c). Here as well as in Figs. 3–5 solid curves show the total DOS per formula unit, dotted curves show the partial T d , and dashed curves the U $5f$ DOS per atom.

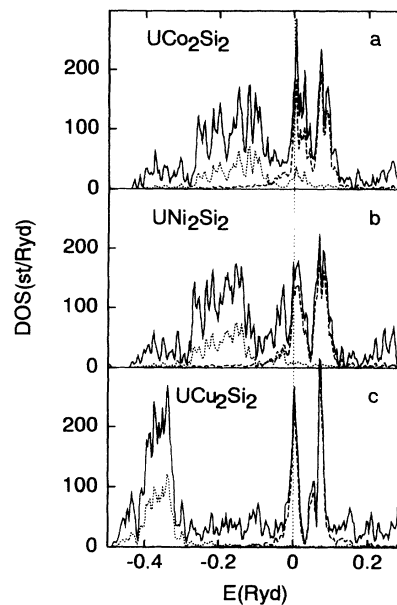


FIG. 3. DOS of nonmagnetic UCo_2Si_2 (a), UNi_2Si_2 (b), and UCu_2Si_2 (c). For comments on the line styles see the caption of Fig. 2.

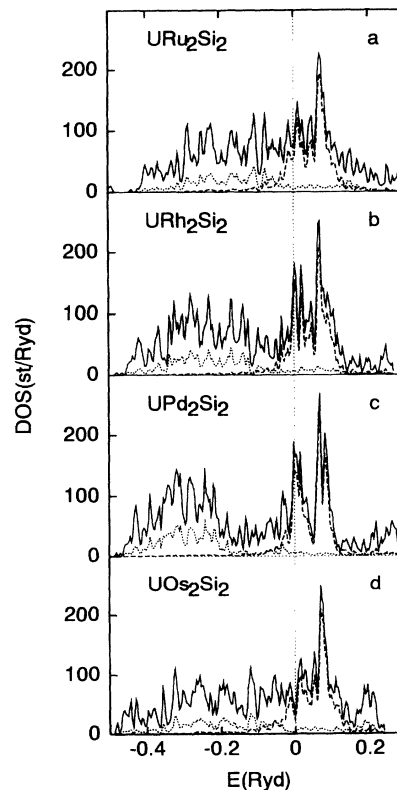


FIG. 4. DOS of nonmagnetic URu_2Si_2 (a), URh_2Si_2 (b), UPd_2Si_2 (c), and UOs_2Si_2 (d). For comments on the line styles see the caption of Fig. 2.

ting of the f states is caused by the spin-orbit interaction. The splitting of the $5f$ states into two groups is still clearly seen in UNi_2Si_2 , UCo_2Si_2 , UPd_2Si_2 , and URh_2Si_2 , although the increasing f - d hybridization leads to an additional structure of the peaks, i.e., a less pronounced minimum between the two groups of $5f$ states and decreasing height of the DOS, especially for the low-energy group of states. With further increase of the f - d hybridization (UFe_2Si_2 , URu_2Si_2 , UOs_2Si_2) the first high $5f$ peak disappears and is replaced by a rich structure of smaller peaks. In the second less hybridized part of the $5f$ states the high peak is still observed but no minimum separating this peak from the other $5f$ states can be seen. Mixing of the f and d states increases even more for UMn_2Si_2 and UCr_2Si_2 where we obtain very strong overlapping d and f bands.

For all compounds where the hybridization is weak (UCu_2Si_2 , UNi_2Si_2 , UCo_2Si_2 , UPd_2Si_2 , and URh_2Si_2) we obtain, in agreement with experiment, the U atom to be magnetic and the T atom to be nonmagnetic. Three compounds of intermediate mixing strength (UFe_2Si_2 , URu_2Si_2 , UOs_2Si_2) are found to be nonmagnetic. A further increase of hybridization (UMn_2Si_2 , UCr_2Si_2) leads in our calculations to both atoms to be magnetic. (Experimentally the presence or absence of a moment on the Cr atom is still not clear.²)

The data shown in Figs. 2–4 allow us to relate the magnetic state of the U and T atoms to the value of the DOS at the Fermi energy (E_F). For all ten compounds the Fermi level lies in the low-energy part of the $5f$ bands and leads to nearly the same number of occu-

ried $5f$ states, namely, 2.8–2.9 per U atom (Table III). For all compounds with weak d - f hybridization we obtain a high $5f$ DOS and low d DOS at E_F . For UFe_2Si_2 , URu_2Si_2 , and UOs_2Si_2 the d DOS at E_F is still low but now the Fermi level lies in the minimum of the U DOS. These minima are caused by the hybridization of d and f bands leading to additional structure of the DOS. Finally, for UMn_2Si_2 and UCr_2Si_2 both $5f$ DOS and d DOS have a substantial value.

Quantitatively, the relation between the DOS at E_F and the magnetic state of the U and T atoms can be established using the Stoner criterion¹⁷ which defines the instability of the nonmagnetic state with respect to the formation of the ferromagnetic state. Our calculations for all magnetic compounds showed that the magnetic state, i.e., the presence or absence of the magnetic moment of both U and T atoms, is independent of the assumed ferromagnetic or antiferromagnetic configuration. Thus, for the present qualitative discussion, the use of the ferromagnetic Stoner criterion is justified. A full account of the different magnetic configurations will be published elsewhere. Our estimates of the Stoner parameter from the coefficient of proportionality between the exchange splitting of the U $5f$ states and the magnetic moment of the U atom, as well as the densities of states at the Fermi energy and the Stoner product, are collected in Table IV together with analogous information for the transition elements. The Stoner parameters for $3d$ and $4d$ elements were taken from Ref. 18. For Os the calculations were done by Mavromaras.¹⁹ The spin-polarized calculations actually do yield magnetic states in those cases where

TABLE IV. Application of the Stoner criterion to the U and T atoms in UT_2Si_2 compounds. I_f and I_d are the Stoner parameters for the U $5f$ states and T d states; $N_f(E_F)$ and $N_d(E_F)$ are corresponding DOS per atom. F denotes that the Stoner product is larger than unity and the Stoner criterion of magnetic instability is fulfilled; N denotes that the product is less than unity and the criterion is not fulfilled.

	5f states of U			d states of T				
	I_f (mRy)	$N_f(E_F)$ (1/Ry)	$I_f N_f(E_F)$	I_d (mRy)	$N_d(E_F)$ (1/Ry)	$I_d N_d(E_F)$		
UCr_2Si_2	17	84	1.4	F	28	28	0.8	N
UMn_2Si_2	16	98	1.6	F	30	37	1.2	F
UFe_2Si_2	17	17	0.3	N	34	4	0.1	N
UCo_2Si_2	17	117	2.0	F	36	15	0.5	N
UNi_2Si_2	18	169	3.0	F	37	9	0.3	N
UCu_2Si_2	17	199	3.4	F	27	3	0.1	N
URu_2Si_2	17	57	0.97	N	22	7	0.2	N
URh_2Si_2	17	132	2.2	F	24	8	0.2	N
UPd_2Si_2	17	155	2.6	F	25	5	0.1	N
UOs_2Si_2	17	19	0.3	N	22	3	0.1	N

the Stoner product is larger than 1. The only exception is the compound UCr_2Si_2 . Here the Stoner criterion is fulfilled for the U atom, and for the Cr atom the product $IN(E_F)$ is high having a value of 0.8 but below unity; i.e., Cr should be nonmagnetic. Still, in spin-polarized calculations both atoms develop a magnetic moment. This disagreement can be explained because in the simple form of the Stoner criterion used here any hybridization between the U and T states is neglected. But hybridization is strong in UCr_2Si_2 .

Now we will comment on some interesting features in URu_2Si_2 and UFe_2Si_2 . The system URu_2Si_2 is the only compound where we did not achieve agreement with experiment concerning the magnetic state of the U atom. Our calculations fail to give a small U moment of $0.04 \mu_B$. Because of the ten compounds it is only URu_2Si_2 which possesses heavy-fermion and superconducting properties, we postulate that the small U magnetic moment as well is caused by many-body quantum interactions that our LSDF calculations can only describe in an approximate way. Furthermore, we note that in our calculations for URu_2Si_2 there is another feature that makes it different from all the other compounds. This is the fact that URu_2Si_2 lies on the boundary between magnetic and nonmagnetic states as can be seen from its Stoner product being very close to unity. On the present level of understanding of the problem we cannot say how important this property could be for the physics of URu_2Si_2 . For example, the maximum in the temperature dependence of magnetic susceptibility²⁰ could have the same spin-fluctuation properties as almost magnetic Pd,²¹ Good agreement²² between our calculations and the photoemission experiment by Grassmann²³ for URu_2Si_2 , URh_2Si_2 , and UPd_2Si_2 allows us to ascertain that also in the case of URu_2Si_2 the band structure calculations give reliable information about the energy positions of the T d and U f bands.

The only previous LSDF calculations for these compounds known to us are those of Norman *et al.*²⁴ for URu_2Si_2 and nonrelativistic calculations for hypothetical ferromagnetic URu_2Si_2 and URh_2Si_2 in Ref. 25. In these two calculations URu_2Si_2 was found to be magnetic. We treat this difference in the calculational results as a confirmation of the vicinity of URu_2Si_2 to a nonmagnetic-magnetic instability point which depends sensitively on the volume and the crystal structure. In fact, numerical experiments done by us by increasing the lattice constants a and c by 2% and keeping the same z (Table I) result in magnetic URu_2Si_2 . Details of the crystal structure concerning the parameter z were not reported in Refs. 24, 25; thus this could be the reason for the differences in the calculated results.

The case of UFe_2Si_2 is also exceptional since of all the six compounds where T is a 3d transition element it is the only one which possesses a nonmagnetic U atom. In Fig. 2(c) we see that the nonmagnetic state of the U atom is determined by the position of the Fermi level in the deep and narrow minimum of the DOS. This coincidence of the minimum and E_F is a combined effect of many factors such as the crystal structure, the number of electrons, and the hybridization strength. In this circum-

stance the nonmagnetic state of UFe_2Si_2 could be called “accidental” and could thus never be predicted without a band structure calculation. Note that UMn_2Si_2 and UCr_2Si_2 also have a deep minimum somewhat below E_F but at the Fermi energy the DOS is rather high which causes not only the U atom but also the T atom to be magnetic.

Next we show in Fig. 5 the results of some model calculations which demonstrate unambiguously the role of the hybridization of the 5f states with the states of other atoms concerning the formation of the nonmagnetic state of UFe_2Si_2 . By setting to zero the corresponding elements of the secular matrix, we carried out calculations of the DOS without accounting for (a) the spin-orbit interaction [Fig. 5(b)], (b) the hybridization of the 5f states and the states of the Fe atom [Fig. 5(c)], and (c) the hybridization of the 5f states with the states of the Fe and Si atoms [Fig. 5(d)]. We see that “turning off” the spin-orbit interaction does not increase the value of the DOS at E_F and therefore does not change the magnetic state of the compound. However, removing the 5f-Fe hybridization changes the picture drastically. In this case we obtain a very high DOS at E_F [Fig. 5(c)] and, on the

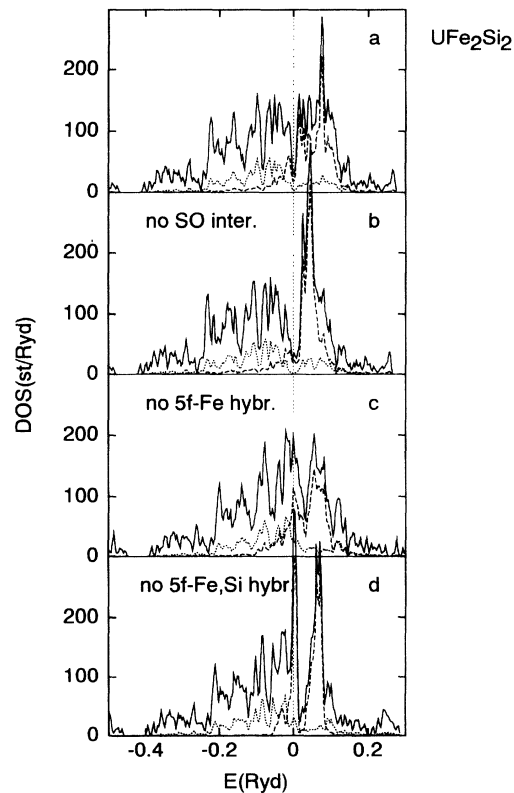


FIG. 5. DOS of nonmagnetic UFe_2Si_2 . (a) Normal calculation, (b) neglect of spin-orbit coupling, (c) neglect of the hybridization between the U 5f states and Fe states, (d) neglect of the hybridization between the U 5f states and Fe and Si states. For comments on the line styles see the caption of Fig. 2.

basis of the Stoner criterion, the compound should be magnetic. Removing the $5f$ -Si hybridization as well we obtain a further increase of the DOS at E_F . This strong influence of the $5f$ -Si hybridization is rather remarkable because the Si bands are very broad and, hence, the partial Si DOS is very low in this energy region compared to that of the $5f$ states.

Referring to the calculated values of the spin and orbital moments mentioned above, we note that they do not reproduce the value of the U magnetic moment estimated from neutron diffraction experiments (see, e.g., the data collected in Ref. 6). From preliminary calculations we know, however, that agreement can be achieved by including into the calculational scheme an additional orbital polarization of the $5f$ states proportional to the value of the orbital atomic moment.²⁶ This additional interaction leads to an increased value of the orbital moment accompanied by changes in the internal structure of the U $5f$ bands that, nevertheless, do not change the position of the U $5f$ bands relative to the $T d$ states and E_F . These new calculations are underway and will be reported in a later publication. A surprising feature of the present results for spin and orbital magnetic moments (Table II) is that they are in good agreement with measurements of the saturation magnetization for the ferromagnetic compounds UMn_2Si_2 and UCu_2Si_2 carried out by Buschow and Mooij.³ The reported experimental values are $3.91\mu_B$ and $1.0\mu_B$ per chemical formula unit to be compared with our theoretical values $3.88\mu_B$ and $0.74\mu_B$, respectively.

III. CONCLUSIONS

Summing up, we state that our calculations show marked and characteristic differences in the electronic structures of the members of the series of compounds UT_2Si_2 ($T=Cr, Mn, Fe, Co, Ni, Cu, Ru, Rh, Pd, Os$); they lie in the different relative energy positions, and correspondingly in the hybridization strengths, of the $T d$ and the U f states. We obtain very good agreement between experiment and theory concerning the presence or absence of the magnetic moment on both the T and U atoms. We show explicitly that this property is determined by the relative position and hybridization of the U and T states. Our calculation does not correctly reproduce the experimentally observed small magnetic moment in URu_2Si_2 . We propose that this small magnetic moment as well as the low-temperature heavy-fermion behavior of URu_2Si_2 and its superconductivity are determined by many-body quantum interactions which enter LSDF theory in a simplified form only. However, the present discussion allows us to conclude that the *ab initio* LSDF theory rather than a correlated impurity picture supplies a reliable basis for studying more complicated physical effects determined by many-body interactions.

ACKNOWLEDGMENTS

We thank Dr. C. Geibel for useful discussions. The work was supported by the Deutsche Forschungsgemeinschaft through SFB 252 Darmstadt, Frankfurt, Mainz.

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