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Theoretical aspects of the magnetism in the ferromagnetic $A\text{Fe}_2$ systems (A = U, Np, Pu, and Am)

Olle Eriksson
Condensed Matter Theory Group, Institute of Physics, University of Uppsala, Box 530, S-75121 Uppsala, Sweden

M. S. S. Brooks
Commision of the European Communities, Joint Research Centre, European Institute for Transuranium Elements, Postfach 2340, D-7500 Karlsruhe, Federal Republic of Germany

Börje Johansson
Condensed Matter Theory Group, Institute of Physics, University of Uppsala, Box 530, S-75121 Uppsala, Sweden

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We report on spin-polarized energy-band calculations for the cubic Laves-phase systems UFe_2, NpFe_2, PuFe_2, and AmFe_2. The calculations were performed with the local-density approximation for the exchange and correlation potential together with a term that shifts the one-electron eigenvalues and takes into account the different interelectronic repulsions for electrons with different $5f$ magnetic quantum numbers. The spin-orbit interaction was also included in the band Hamiltonian. Thus the parameter-free calculations incorporate Hund's first, second, and third rules. The magnetism in the first three compounds was found to be dominated by a large orbital contribution coupled antiparallel to the spin moment. In the calculations for AmFe_2, the $5f$ electrons were treated as core electrons. The magnetism was here found to behave very much like the magnetism of similar rare-earth Laves-phase compounds.

I. INTRODUCTION

The magnetic properties of the cubic Laves-phase $A\text{Fe}_2$, (A=U, Np, Pu, and Am) systems are complex in that the magnetic moments have substantial contributions both from the Fe and the A atoms. In connection with the interpretation of these magnetic properties it has been debated whether the $5f$ electrons in these systems form band states or if they are localized. From magnetization measurements, magnetic anisotropy studies, and neutron-scattering experiments, it has been argued that the $5f$ electrons in UFe_2 are delocalized whereas they are localized in NpFe_2 and PuFe_2.[1-4] Neutron experiments for AmFe_2 indicated a small moment at the Am site and this led the authors to suggest that the valence of the Am ion is of mixed type in this compound.[1] Later neutron experiment gave further support to the picture of delocalized $5f$ electrons in UFe_2,[5] whereas the localized $5f$ state in PuFe_2 was questioned.[6]

UFe_2 and PuFe_2 have also been studied by means of photoelectron spectroscopy, and both showed a pronounced structure at the Fermi level that extended some 2–3 eV below and which originates from $5f$ states.[7,8] Furthermore, for UFe_2 the linear specific-heat coefficient was measured to be as high as 45 mJ/mol K^2 (Ref. 9) and the pressure derivative of the magnetic moment, $d\ln\mu/dP$, was found to be $-4.8\ \text{Mbar}^{-1}$ (Ref. 10).

The first attempt to calculate the electronic structure of actinide Laves-phase compounds was not based upon band-structure calculations but upon single-site scattering theory where only the scattering phase shifts were calculated.[11] Although the conclusions to be drawn from such calculations, which contain no hybridization and are not self-consistent, are necessarily somewhat vague, they did show that the $5f$ resonance was located at the top of the $3d$ resonance—as would be expected.

The first energy-band calculations were made for UFe_2 and UCo_2 (Ref. 12) and concentrated upon calculations of the lattice constant, the onset of ferromagnetism in a detailed multiband Stoner theory, and the magnitude of the moment in self-consistent spin-polarized calculations. All indications were that the $5f$ electrons were correctly treated by self-consistent energy-band calculations within the local approximation to the density-functional theory (LSDA). However, although the calculated iron moment of $0.8\mu_B$ compared with the measured moment of $0.6\mu_B$ was not extravagantly large, the present authors[12] were made suspicious of their own results by the calculated magnitude of the uranium moment, $-0.6\mu_B$/atom which they compared with the measured uranium moment of $0.06\mu_B$/atom.[1,13]

Subsequently, other authors made spin-polarized calculations for UFe_2 (Ref. 14) and it was claimed that the calculated total moment was in reasonable agreement with experiments. However, no examination of the uranium form factor was made and the reservations expressed above about the results of our own earlier calculations apply to these calculations also.

The present authors then made a detailed examination of a series of experimental results and compared them with self-consistent spin-polarized calculations with spin-orbit coupling included.[15,16] It was found that the spin-induced orbital contribution to the magnetic moment at the uranium site was as large as the spin contribution. From this they were able to predict that the magnetic form factor should be anomalous. Such an anomalous form factor has subsequently been measured.[5]

Recently it has been shown that a relativistic generalization of the Stoner criterion for the onset of itinerant ferromagnetism predicts $PuFe_2$ to order magnetically whereas, for instance, $PuCo_2$ should be paramagnetic.[17] This agrees with experiments and gives a clear indication that it is the Fe atom which is responsible for the ferromagnetism and that via hybridization induces a moment on the A site in $PuFe_2$ and possibly also in the other AFe_2 systems.

In this paper we report on band-structure calculations for the AFe_2 systems (A=U, Np, Pu, and Am). The calculations were performed in the same manner as described elsewhere,[18,15] utilizing the linear muffin-tin orbital method (LMTO), with the von Barth–Hedin exchange and correlation potential[19] and with the frozen-core approximation. In the treatment, the $5f$ electrons were considered as band electrons for UFe_2, $NpFe_2$, and $PuFe_2$. This was done to investigate to what extent such an approach can explain the magnetic properties of these systems and to provide additional insight to their complex magnetic and electronic properties. In this study we have also included energy eigenvalue shifts for $5f$ states with different magnetic quantum numbers m_l. This takes into account the different intra-atomic Coulomb interactions between electrons with different magnetic quantum numbers. Thus, the energies of these orbitals are split, implementing Hund's second rule in the energy-band calculation.[20] However, for $AmFe_2$ we have treated the $5f$ electrons as core states with a $5f^6$ configuration, because this is the most likely electronic configuration of the Am ion in $AmFe_2$.

The electronic structure obtained from normal band calculations is presented in Sec. II. Section III discusses the calculated magnetic moments from a standard spin-polarized band calculation without spin-orbit coupling. In Sec. IV we present the theoretical magnetic moments obtained when the spin-orbit coupling has been included and Sec. V gives the results from calculations with the above-mentioned magnetic quantum-number splittings, used together with the spin-orbit coupling. In Sec. VI we present calculations of magnetic form factors and Sec. VII contains conclusions.

II. ELECTRONIC STRUCTURE

The calculated paramagnetic density of states (DOS) for UFe_2, $NpFe_2$, and $PuFe_2$ are depicted in Fig. 1. Here we see that, just as for UFe_2,[12,14] the electronic structure is composed of two dominating features, namely the $3d$ and the $5f$ partial DOS. These two features are relatively close in energy, with the $5f$ resonance at higher energies than the $3d$ resonance. The Fermi level E_F lies in the middle of these two bands in a region with a high density of states. Therefore, we calculate the value of the Stoner product to be larger than one for all three systems and we expect them to order magnetically. In Fig. 1 we also notice that the $5f$-band width becomes more narrow as we move across the series, which is an effect of incomplete screening of the extra added electron when the nuclear charge increases. This, in turn, increases the local exchange integrals[21] at the A site and, as we will see below, this influences the magnetic properties in a quite dramatic way at the Fe site also.

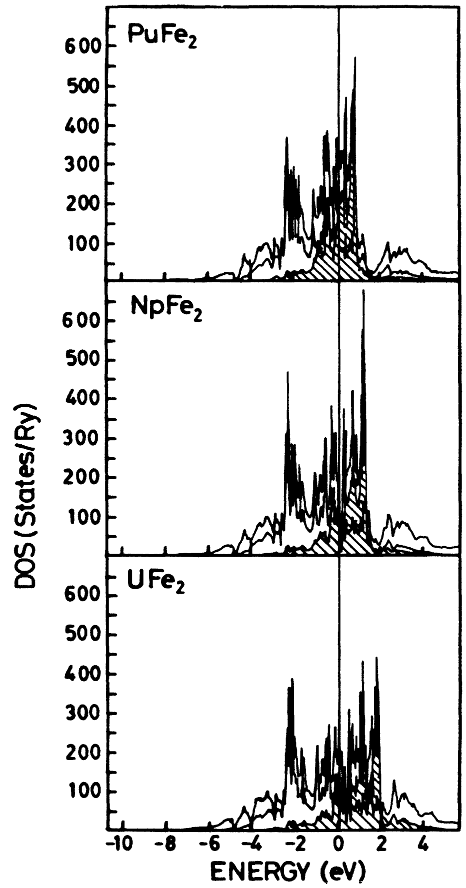

FIG. 1. Calculated DOS for UFe_2, $NpFe_2$, and $PuFe_2$. The upper curve in each panel is the total DOS. The lower curves in each panel are the Fe d and U f partial DOS. The area below the U f DOS is dashed.

In Fig. 2 we have plotted the DOS for the paramagnetic state of $AmFe_2$. This calculation was performed with the $5f$ electrons treated as core electrons. The DOS for $AmFe_2$ is very similar to that of the rare-earth iron Laves-phase intermetallics,[22] and it is dominated by the Fe $3d$ bands which are pinned at the Fermi level. The Stoner product is larger than one also for this system and therefore a ferromagnetic state is expected to be stable. Since these calculations did not included the $5f$ states we conclude that the magnetism of the AFe_2 compounds is driven by the Fe atoms.

III. MAGNETIC MOMENT FROM A STANDARD BAND CALCULATION

We first made normal self-consistent spin-polarized calculations allowing for different potentials for the spin-up and spin-down electrons. The total and partial magnetic

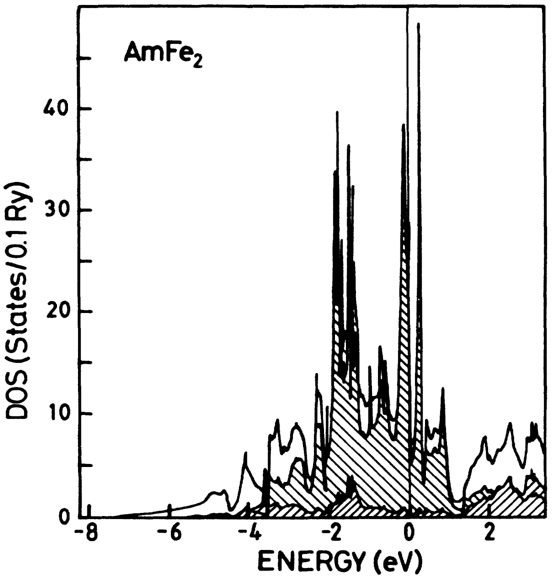

FIG. 2. Calculated DOS for AmFe$_2$. The upper curve is the total DOS. The Fe d partial DOS is dashed from left to right and the Am d partial DOS is dashed from right to left. In these calculations the $5f$ electrons are considered as core electrons.

moments from these calculations are given in Table I. Here we see that the Fe $3d$ partial moments compare quite well with the moments obtained from neutron-scattering measurements for all four compounds (Table III). Superficially the magnetic moment on the Am site in AmFe$_2$ compares quite well with the measured moment (Table III) but we find it to be due to the $6d$ electrons whereas the measured moment is—due to its form factor—attributed to $5f$ electrons.[1] Furthermore, the agreement between experiment and theory for the A moment is bad for the compounds UFe$_2$, NpFe$_2$, and PuFe$_2$. A reason for this could be the orbital contribution to the magnetic moment, a well-known feature of magnetism in actinide systems.[15]

Neglecting for the moment the mentioned discrepancy for the actinide moment, it is interesting to see that the trend of the experimental Fe moments in these systems is well reproduced by theory although the absolute values deviate somewhat from the experimental data. From Table I we notice that the $3d$ and the $5f$ moments are calculated to be antiparallel. The reason for this has already been explained in detail in a study of rare-earth intermetallic compounds[23,24] and is due to the fact that the hybridization between the $3d$ and the $5f$ states for the majority bands is weaker than for the minority bands.

TABLE I. Calculated moments (in units of μ_B) from a normal spin-polarized calculation. μ_{Fe} is the total Fe moment, $\mu_{\mathrm{Fe}\ d}$ is the Fe d moment, μ_A is the total A moment, and $\mu_{A\ f}$ is the A f moment.

	UFe$_2$	NpFe$_2$	PuFe$_2$	AmFe$_2$
μ_A	−0.61	−0.84	−4.59	−0.30
$\mu_{A\ f}$	−0.51	−0.73	−4.15	
μ_{Fe}	0.77	0.69	1.25	1.81
$\mu_{\mathrm{Fe}\ d}$	0.75	0.67	1.25	1.84

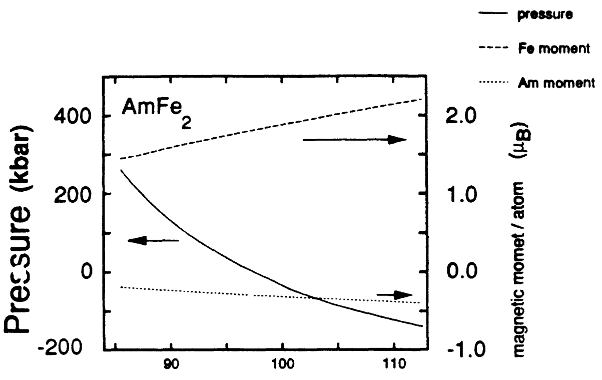

FIG. 3. Spin-polarized equation of state for AmFe$_2$. Also plotted are the partial magnetic moments for the Fe and Am atoms.

We have also calculated the equation of state for AmFe$_2$ and this is shown in Fig. 3. Here, also, the individual Fe and Am magnetic moments are shown as a function of volume. The agreement between theory and experiment concerning the lattice constant is found to be good.

IV. MAGNETIC MOMENT WHEN INCLUDING SPIN-ORBIT INTERACTION

As stated above it is known that the orbital contribution to the magnetism in actinide compounds with delocalized $5f$ electrons can be substantial.[15,16,25,26] Therefore, we have calculated the orbital contribution to the magnetic moments for UFe$_2$, NpFe$_2$, and PuFe$_2$. This was done by including the spin-orbit interaction in the calculation in the same way as described in Ref. 15. Both the Fe and the A moments obtained from these calculations are listed in Table II. Here we see that the Fe spin moment for UFe$_2$ is influenced very little by this in-

TABLE II. Calculated moments (in units of μ_B) from a spin-polarized calculation with spin-orbit interaction. $\mu_{S\text{-Fe}}$ is the total Fe spin moment, $\mu_{S\text{-Fe}\ d}$ is the Fe d spin moment, $\mu_{S\text{-}A}$ is the total A spin moment, and $\mu_{S\text{-}A\ f}$ is the A f spin moment. Also given are the orbital contributions for the Fe and the A site, denoted $\mu_{L\text{-Fe}}$ and $\mu_{L\text{-}A}$, respectively. The sum of the spin and the orbital moments are denoted $\mu_{\mathrm{tot}\text{-}A}$ and $\mu_{\mathrm{tot\text{-}Fe}}$ for the A and the Fe site, respectively.

	UFe$_2$	NpFe$_2$	PuFe$_2$
$\mu_{S\text{-}A}$	−0.71	−1.38	−3.89
$\mu_{S\text{-}A\ f}$	−0.58	−1.17	−3.48
$\mu_{S\text{-Fe}}$	0.75	0.88	1.35
$\mu_{S\text{-Fe}\ d}$	0.73	0.87	1.34
$\mu_{L\text{-}A}$	0.47	0.70	1.45
$\mu_{L\text{-Fe}}$	0.07	0.09	0.08
$\mu_{\mathrm{tot}\text{-}A}$	−0.24	−0.68	−2.44
$\mu_{\mathrm{tot\text{-}Fe}}$	0.82	0.97	1.43
$\mu^{\mathrm{Fe}}_{\mathrm{expt}}$ (Ref. 1)	0.59	1.35	1.47

clusion. However, for $NpFe_2$ and $PuFe_2$, the iron spin moment increases by about 10–30 % and now the agreement between theory and experiment is even better than for the normal spin-polarized calculation. It is also clear that the A spin moment changes dramatically especially for $PuFe_2$. The A moment now has a large orbital component and again $PuFe_2$ shows the largest effect. The orbital contribution to the Fe moment in these systems is found to be quite small, with a magnitude of about $0.08\mu_B$ and parallel to the Fe spin moment. This is similar to the orbital moment for other Fe systems.[23,27] The overall agreement between theory and experiment here is quite good for UFe_2 (as we have reported earlier[16]), but for $NpFe_2$ and $PuFe_2$ the agreement for the A moment is poor. Even the calculated sign of the A moment is incorrect. However, for all three systems we calculated a magnetic moment on the Fe site that agrees quite well with the experimental data, being substantially smaller than the Fe moment in $AmFe_2$ and similar rare-earth iron Laves-phase systems. The Fe magnetic moments increase steadily across the series from UFe_2 to $AmFe_2$, a fact that seems puzzling since the change in volume for the three systems cannot alone explain this trend.

The effect of volume on the magnetic moments in these types of systems is quite small as we have shown earlier[16] and as can also be seen from the change of the iron moments in $AmFe_2$ with volume (Fig. 3). In order to illuminate the reason for the trend of the Fe moments in these systems we plot in Fig. 4 the DOS for UFe_2, $NpFe_2$, and $PuFe_2$ obtained from the normal spin-polarized calculations. Here we see that the spin splitting of those bands which are dominated by Fe $3d$ character is almost constant for the three compounds. However, due to hybridization with the A $5f$ bands there is a strong $5f$ wave-function admixture into these bands. If this admixture is equal, or almost equal, for the spin-up and the spin-down bands there will be a reduction of the Fe moment. This can be understood by approximating the Fe $3d$ band by a rectangular DOS whereby the magnetic moment on the Fe atom is simply

$$\Delta_{\text{exchange}} \frac{D_{\text{Fe}}}{2} \, , \qquad (1)$$

where Δ_{exchange} is the spin splitting and D_{Fe} is the Fe $3d$ state density. If the $5f$ wave admixture into this rectangular DOS increases, the Fe partial DOS in Eq. (1) will decrease and so will the Fe moment. However, this is, to some extent, compensated by the fact that the hybridization is stronger between the spin-down (minority) bands than for the spin-up (majority) bands. For UFe_2 the first effect dominates and thus there will be a dramatic reduction of the magnetic moment compared to that of similar systems with little or no $5f$ admixture into the "$3d$ bands." However, for $NpFe_2$ and especially $PuFe_2$ the local $5f$ exchange interactions at the A site splits the $5f$ band, so that the hybridization between the $3d$ and $5f$ states will now become quite strong for the spin-down band. This decreases the number of minority $3d$ electrons. Hence, when this second compensating effect will start to become substantial, the Fe moment will increase. This is what happens in $NpFe_2$ and even more so in $PuFe_2$. Finally in $AmFe_2$ there is no, or little, $5f$ admixture into the $3d$ band and the Fe moment is therefore large in this compound.

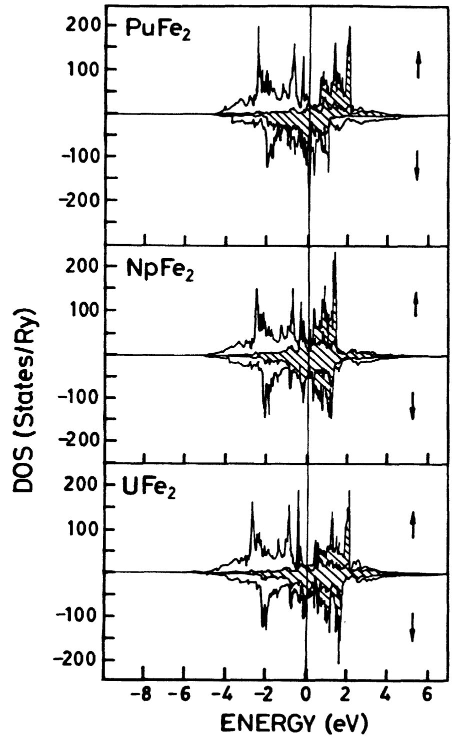

FIG. 4. $3d$ and $5f$ partial DOS for UFe_2, $NpFe_2$, and $PuFe_2$ from a spin-polarized calculation. The $5f$ DOS is dashed from left to right and the arrows indicate the spin-up and spin-down DOS.

This explains quite well the development of the Fe moments through the series of compounds. However, the A moments for $NpFe_2$ and $PuFe_2$ are still not reproduced by our calculations, which therefore indicates that some important physical interactions are lacking. The most apparent case is $PuFe_2$, and in the next section we will describe an attempt to take these residual interactions into account.

V. ORBITAL SPLITTING AND MAGNETIC MOMENTS

In narrow-band systems all Hund's rules must be considered as is certainly the case in the localized regime. Although the interactions leading to Hund's second rule are conventionally neglected in energy-band calculations they are crucial in atoms. Since we are dealing with very narrow-band systems and our calculations indicate that the induced-orbital moments are too small, we are led to suspect that it is these interactions which are important in determining the magnetic moments. The Hund's rules mean that not only the spin moment should be maximized but also the orbital moment. In the limit of completely localized states the ground state of the atomic f^n configuration can be obtained from a vector model[28] involving interactions of type $\overline{s}_i\overline{s}_j$ and $\overline{l}_i\overline{l}_j$ ($\overline{s}_i$ and $\overline{l}_i$ are the spin and angular momenta for the ith electron within the f^n configuration, respectively). By replacing the interaction energy of the form $-\sum_{i,j}\overline{s}_i\overline{s}_j$ with the mean-field approximation $(-\sum_i s_i^z)(\sum_j s_j^z)$ one obtains an energy $-\frac{1}{4}IM_s^2$ (M_s=spin magnetization$=2S_z$ and I=Stoner exchange parameter) which is the Stoner expression for the spin-polarization energy.[29] Hence, the Stoner energy is a function of the spin and the spin-up and spin-down bands are split rigidly. For the orbital polarization we follow an analogous route. We replace $-\frac{1}{2}\sum_{i,j}\overline{l}_i\overline{l}_j$, which occurs in the energy expression of the ground state of an atom as a function of occupation number with $-\frac{1}{2}(\sum_i l_i^z)(\sum_j l_j^z)$. Hence, we obtain a term proportional to $-E^3(L^2/2)$ where E^3 is the Racah parameter[30,28] and L is the total angular momentum. From this we obtain the corresponding one-electron eigenvalue shift $(-E^3Lm_l)$ for the m_l state. Thus, the spin-up or spin-down manifolds are split when the total orbital moment is nonzero. E^3 now plays the analogous role to the Stoner I. We have previously performed calculations with these eigenvalue shifts together with the exchange-correlation and spin-orbit interactions for UCoAl,[31] $NpAl_2$,[32] $NpOs_2$,[33] $NpCo_2$,[34] and obtained good agreement with experiment concerning the magnetic and bonding properties. We have also applied this to the γ-α transition in Ce.[20] Here we obtained good agreement with experiment, both as regards the magnetic properties as well as the volume collapse accompanying the γ-α transition in Ce.

Here we apply the above-mentioned shifts, together with the exchange-correlation potential as well as the spin-orbit interactions to the electronic structure of UFe_2, $NpFe_2$, and $PuFe_2$. Thus, we have done parameter-free calculations that include Hund's first, second, and third rules. The self-consistently calculated E^3 values are 3.5, 4.0, and 4.6 mRy for UFe_2, $NpFe_2$, and $PuFe_2$, respectively. The magnetic moments from these calculations are listed in Table III together with available experimental data. In order to compare our theoretical results with the neutron-scattering measurements for UFe_2, $NpFe_2$, and $PuFe_2$ we have also listed the A 5f moments. As can be seen from Table III the experimental A 5f moments and theory now compare very well. Also, the theoretical value for the moment compares well with the moment obtained from Mössbauer experiments for $NpFe_2$. In all these systems we find a situation where the A 5f orbital moment is larger in size and antiparallel to the A 5f spin moment and therefore the 5f moment becomes parallel to the iron moment. From Table III we also notice that there is a moment on the A site that originates from the 6d orbitals, which is parallel to the (negative) 5f spin moment. The theoretical values for the total moments compare relatively well with magnetization measurements. Due to anisotropy it is likely that the magnetization measurements have not been driven to saturation. From this point of view it is satisfactory to note that the calculated magnetic moments are larger than those obtained experimentally.

In Figs. 5 and 6 we summarize the calculated moments

TABLE III. Experimental and theoretical data of the magnetic properties of the AFe$_2$ systems. γ is the coefficient of the linear contribution to the specific heat. μ_0 is the total magnetic moment from magnetization measurements. μ^N denotes the moment from neutron experiments and μ^{hf} that from hyperfine-field measurements. Also given are the theoretical moments, with the same notations as in Table II.

	UFe_2	$NpFe_2$	$PuFe_2$	$AmFe_2$
T_c (K) (Ref. 13)	162	492	564	613
γ (mJ/mol K^2) (Ref. 9)	45			
μ_0 (μ_B) (Ref. 3)	1.11	2.58	2.28	3.13
μ_{Fe}^N (μ_B) (Ref. 1)	0.59	1.35	1.47	1.7
μ_A^N (μ_B) (Ref. 1)	0.06	1.09	0.45	-0.4
μ_A^{hf} (μ_B) (Ref. 2)		0.87		
$d\ln\mu/dP$ (Ref. 10)	-4.8			
$\mu_{A\,f}^N$ (Ref. 5)	0.01		0.39	
$\mu_{\mathrm{Fe}\,d}^N$ (Ref. 6)	0.60		1.73	
$\mu_{\mathrm{theor}}^{\mathrm{tot}}$	1.63	3.25	2.89	3.38
$\mu_{\mathrm{theor}}^{S\text{-}A\ \mathrm{tot}}$ (μ_B)	-1.03	-2.60	-3.55	
$\mu_{\mathrm{theor}}^{S\text{-}A\ f}$ (μ_B)	-0.83	-2.29	-3.19	
$\mu_{\mathrm{theor}}^{L\text{-}A}$ (μ_B)	0.88	3.49	3.52	
$\mu_{\mathrm{theor}}^{\mathrm{tot}\text{-}A}$ (μ_B)	-0.15	0.89	-0.03	-0.3
$\mu_{\mathrm{theor}}^{\mathrm{tot}\text{-}A\ f}$ (μ_B)	0.05	1.20	0.33	
$\mu_{\mathrm{theor}}^{S\text{-Fe}}$ (μ_B)	0.82	1.09	1.38	
$\mu_{\mathrm{theor}}^{L\text{-Fe}}$ (μ_B)	0.07	0.09	0.08	
$\mu_{\mathrm{theor}}^{\mathrm{tot\text{-}Fe}}$ (μ_B)	0.89	1.18	1.46	1.84
C_2 (from f moment)	18	3	11	
C_2 (expt.) (Refs. 5 and 6)	23		6	

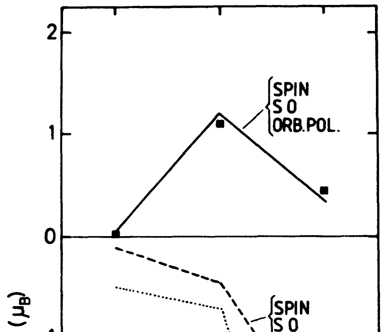

FIG. 5. Calculated (lines) and experimental (squares) 5f moments for UFe_2, $NpFe_2$, and $PuFe_2$. The magnetization of Fe is in the positive direction.

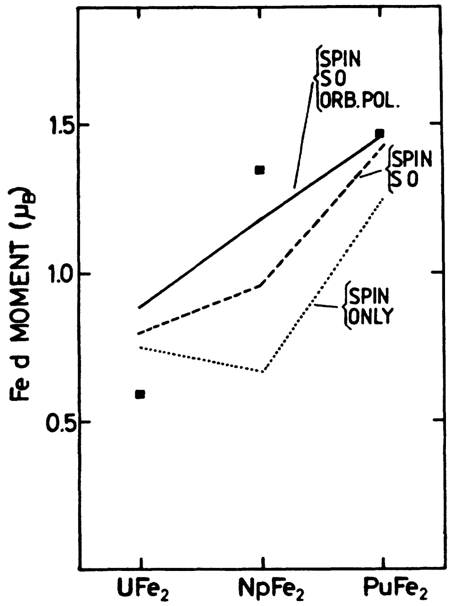

FIG. 6. Calculated (lines) and experimental (squares) $3d$ moments for UFe$_2$, NpFe$_2$, and PuFe$_2$.

for the $5f$ electrons and the $3d$ electrons, respectively. The importance of including the spin orbit and the orbital polarization is most clearly seen for the $5f$ moments (Fig. 5). This should, of course, be expected since it is $5f$ magnetization which is most susceptible to these interactions. However, there is also an indirect influence on the $3d$ moment as is illustrated in Fig. 6.

Also, for AmFe$_2$ do we find that the comparison of the magnetic moments between theory and reported data is quite good. The magnetic moment is here almost entirely located at the Fe site and it is large, just as in the rare-earth iron Laves phases. There is, however, a small moment located at the Am atom which originates from the $6d$ spin density.

VI. MAGNETIC FORM FACTORS

A powerful method for probing the nature of the magnetic moments on an atomic scale is neutron scattering. We have, as a test of the above results, calculated the magnetic form factors for UFe$_2$, NpFe$_2$, and PuFe$_2$, and compared these with the experimental form factors. The calculations of the spherical form factors were done in the dipole approximation[35]

$$f(Q)=\langle j_0\rangle+C_2\langle j_2\rangle \ . \tag{2}$$

Here $Q=4\pi\sin\theta/\lambda$ is the momentum transfer, and $\langle j_n\rangle$ is the average of spherical j_n Bessel function for the spin density. Furthermore, C_2 is the ratio μ_l/μ where μ_l is the orbital moment and μ is the total moment on the site. This ratio is quoted from experiments for the A atoms for UFe$_2$, NpFe$_2$, and PuFe$_2$ in Table III,[5,6] together with our theoretical values. The calculated A form factors are plotted in Fig. 7. For UFe$_2$ and PuFe$_2$ they are characterized by a pronounced maximum for intermediate Q values. This agrees exactly with the observations for UFe$_2$ and PuFe$_2$. It should be emphasized that these form factors are quite different from those derived from

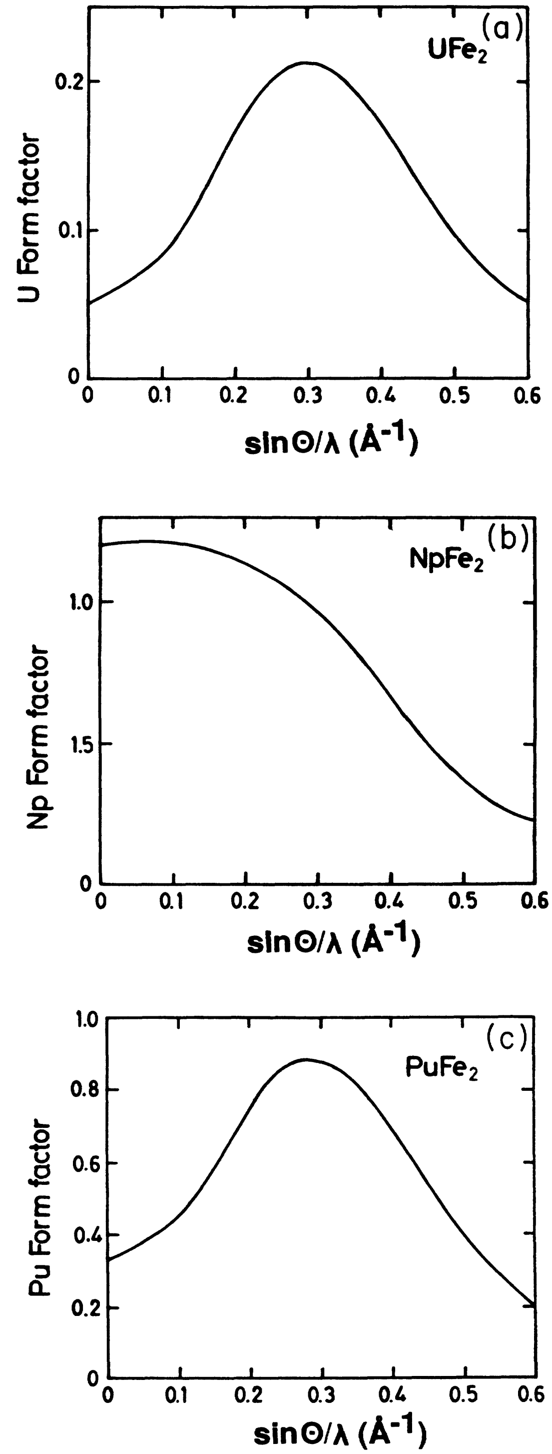

FIG. 7. (a) Magnetic form factor times the magnetic moment for U in UFe$_2$. (b) Magnetic form factor times the magnetic moment for Np in NpFe$_2$. (c) Magnetic form factor times the magnetic moment for Pu in PuFe$_2$.

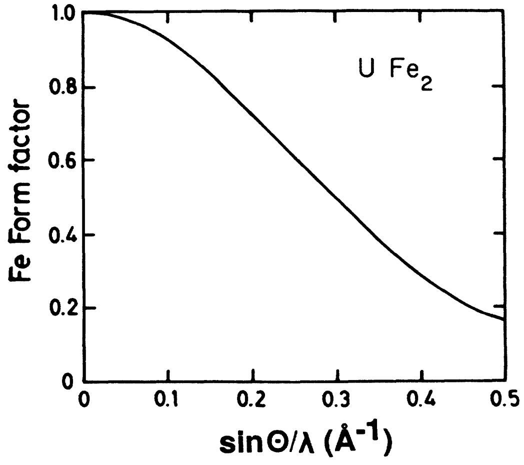

FIG. 8. Magnetic form factor for Fe in UFe$_2$.

either theories using localized $5f^n$ configurations or conventional spin-polarized band states. We are not aware of any published data for the shape of the Np form factor for NpFe$_2$, but we calculated C_2 to be smaller than for the other systems and thus the upturn of the form factor will only be present for very low values of Q and thus very difficult to observe. As stated above the A $5f$ moment obtained from theory and from neutron experiments compare quite well for all three systems. However, since C_2 is inversely proportional to the total moment μ, it becomes a very sensitive quantity in case when the total value of μ is small. This is the case for UFe$_2$ and PuFe$_2$ and therefore the good agreement between theory and experiment for C_2 is gratifying. From Table III we also notice that in the calculations there is a clear difference between the A $5f$ moment and the total A moment. Without commenting upon the validity of deducing the total moment at the Np site from hyperfine field measurements[36] we have listed in Table III Np moments obtained from such data. We see here that the difference in moments between the different experimental techniques (Mössbauer or neutron experiments) is in good accordance with our calculations, when comparing the total A moment to the $5f$ moment, both as regards the general trend ($\mu_{\text{Mössbauer}} < \mu_{\text{neutrons}}$) and also for the absolute values.

We have also calculated the Fe form factor for all four compounds and since they are all very similar we have chosen to plot only the normalized Fe form factor for UFe$_2$. This is shown in Fig. 8 and we find that it behaves much like the Fe form factor in α-Fe.[37]

VII. CONCLUSION

We have demonstrated that the complicated magnetic properties of the actinides in the AFe$_2$ systems are dominated by an orbital contribution for UFe$_2$, NpFe$_2$, and PuFe$_2$. The orbital moment was actually found to be larger in size but antiparallel to the spin moment, which resulted in rather low total actinide moments for UFe$_2$, NpFe$_2$, and PuFe$_2$. The orbital moment was calculated using a potential that accounts for the intra-atomic Coulomb interaction between electrons in orbitals of different magnetic quantum numbers. Since the calculations included exchange, correlation, and spin-orbit interactions, we have performed a parameter-free calculation with Hund's first, second, and third rules taken into account. Only when we include all these terms to the treatment do the A $5f$ moments compare well with the moments derived from neutron-scattering experiments. The calculated magnetic form factors also reproduce the general shape suggested from the experimental data. We have also shown that the $5f$-$3d$ hybridization reduces the magnetic moment of the $3d$ electrons and induces a negative spin moment at the A site. However, we find that local exchange interactions on the A atom in their turn increase the Fe moment and that they become more important as the actinide atomic number increases. The moment on the Fe site is shown to be dominated by the spin contribution and already ordinary spin-polarized calculations yield magnetic moments that agree relatively well with experiments.

The calculations were performed with the $5f$ electrons treated as band states. Thus, the good agreement between theory and experiment for the magnetic properties supports that a delocalized picture is appropriate for UFe$_2$, NpFe$_2$, and PuFe$_2$. Finally we have calculated the magnetic moment and the equation of state for AmFe$_2$, with the $5f$ electrons put into the core. Also, here we find good agreement between theory and experiment, both for the magnetic moment as well as the experimental lattice constant, which therefore supports the view that the $5f$ electrons are localized in this compound.

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