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Neutron diffraction study of magnetic ordering in UPd_2Si_2 , UPd_2Ge_2 , URh_2Si_2 and URh_2Ge_2

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Abstract. Neutron diffraction measurements on polycrystalline samples show that UPd_2Si_2 , UPd_2Ge_2 and URh_2Si_2 become antiferromagnetic below temperatures of 150 K, 140 K and 137 K respectively. The magnetic moment ordering in UPd_2Si_2 and UPd_2Ge_2 is represented by a static longitudinal wave propagating along the c axis of the tetragonal body-centred crystal lattice. The magnetisation wave is polarised in the direction of the c axis: $q = (0, 0, 0.662 \pm 0.010)c^*$ in UPd_2Si_2 and $q = (0, 0, 0.748 \pm 0.010)c^*$ in UPd_2Ge_2 . At 40 K, UPd_2Si_2 exhibits a magnetic phase transition: a commensurate magnetisation wave with $q = (0, 0, 1)c^*$ represents the growth of a simple antiferromagnetic ordering.

URh_2Si_2 is a collinear antiferromagnet with coinciding magnetic and chemical unit cells. The sequence of ferromagnetic sheets piled up along the tetragonal axis is given by $+$ $-$. The values of the root mean square of the magnetic moment on the U ion at 4.2 K are found to be $1.96 \mu_B$, $1.97 \mu_B$ and $1.96 \mu_B$ for UPd_2Si_2 , UPd_2Ge_2 and URh_2Si_2 respectively. The observed magnetic ordering schemes are discussed in terms of the RKKY exchange interaction. Neutron diffraction measurements performed at zero magnetic field show that URh_2Ge_2 is paramagnetic at 4.2 K.

1. Introduction

Magnetic susceptibility measurements performed on polycrystalline samples of UPd_2Si_2 , UPd_2Ge_2 , URh_2Si_2 and URh_2Ge_2 show that they are all antiferromagnetic with Néel temperatures of 148 K, 139 K, 138 K and 8 K respectively (A Zygmunt 1980, private communication). A neutron diffraction study was therefore undertaken to determine their magnetic structures, since we thought this would provide valuable information regarding the nature of magnetic interactions in these compounds.

2. Experimental procedure

Polycrystalline samples of UPd_2Si_2 , UPd_2Ge_2 , URh_2Si_2 and URh_2Ge_2 were obtained by a method which is described elsewhere (see Marazza *et al* 1977, Zygmunt 1977). They were found by x-rays to have single phases with the determined lattice constants listed in table 1.

Table 1. Crystal structure parameters of UMe_2X_2 compounds.

	UPd_2Si_2 I4/mmm	UPd_2Ge_2 I4/mmm	URh_2Si_2 I4/mmm	URh_2Ge_2 P4/mmm
a (Å)	4.097 ± 0.002	4.200 ± 0.006	4.009 ± 0.002	4.155 ± 0.002
c (Å)	10.046 ± 0.010	10.230 ± 0.018	10.025 ± 0.005	9.771 ± 0.005
K scale factor	0.00282 ± 0.00008	0.00356 ± 0.00004	0.00428 ± 0.00005	0.00572 ± 0.00010
x	0.3816 ± 0.0010	0.3830 ± 0.0008	0.3832 ± 0.0010	$x_1 = 0.4041 \pm 0.0017$ $x_2 = 0.1258 \pm 0.0024$ $x_3 = 0.2500 \pm 0.0010$

Neutron diffraction patterns were taken at 4.2 K, 80 K and 300 K using the powder diffractometer DN-500 at reactor EWA in Swierk. The neutron wavelength was 1.324 ± 0.003 Å. Due to the higher absorption of thermal neutrons by the Rh nuclei a special vanadium container was used with its diameter so chosen as to minimise the absorption factor. In addition the counting time was increased in comparison with the palladium compounds. Runs at 4.2 K were performed with the sample immersed in liquid helium.

The temperature variation of magnetic peak heights was measured using a Fe-Fe(Au) thermocouple while the cryostat was warming up slowly.

Table 2. Comparison of the observed and calculated neutron nuclear intensities at 300 K.

hkl	UPd_2Si_2		UPd_2Ge_2		URh_2Si_2		URh_2Ge_2	
	$I_{\text{obs}}K$	I_{cal}	$I_{\text{obs}}K$	I_{cal}	$I_{\text{obs}}K$	I_{cal}	$I_{\text{obs}}K$	I_{cal}
001		—		—		—	3	6
100		—		—		—	2	0
003		—		—		—	27	12
102		—		—		—	22	24
111		—		—		—	23	21
113		—		—		—	34	22
122		—		—		—	40	16
002	21	18	13	8	13	11	50	22
101	31	30	84	87	31	27	21	26
110	48	36	301	292	58	41	117	117
103	414	469	897	893	401	438	2038	571
112		{ 1070		{ 1222		{ 1023		{ 1492
004	1160	{ 90	1219	{ 10	1130	{ 86	90	60
200	689	671	1251	1180	—	—	1146	1107
114		{ 191		{ 577		{ 172		{ 431
202	206	{ 11	588	{ 5	169	{ 6	435	{ 13
105		{ 305		{ 690		{ 315		{ 528
211	307	{ 15	702	{ 42	251	{ 13	18	13
006	5	8	9	20	10	10	519	{ 35
213		{ 383		{ 733		{ 354		{ 479
204	464	{ 154	741	{ 17	536	{ 143	75	106
116		{ 294		{ 213		—	—	—
220	737	{ 357	781	{ 626	—	—	—	—
	$R = 6.5\%$		$R = 3.4\%$		$R = 7.9\%$		$R = 5.8\%$	

3. The crystal structure

The four compounds investigated belong to a ThCr_2Si_2 crystal structure type, common among rare-earth intermetallics. The space group is $I4/mmm$ and the atomic sites are as follows (International Tables of Crystallography 1952):

2U in 2(a):	0, 0, 0	
4Me in 4(d):	$0, \frac{1}{2}, \frac{1}{4}$	$\frac{1}{2}, 0, \frac{1}{4}$
4Si in 4(e):	$0, 0, x$	$0, 0, \bar{x}$
+ body-centering translation.		

Free atomic parameters were determined from the neutron intensities measured at 300 K using a least-squares computer program. Neutron scattering lengths were taken, following Bacon (1972), to be (in 10^{-14} m):

$$b_U = 0.85 \quad b_{Pd} = 0.60 \quad b_{Rh} = 0.59 \quad b_{Si} = 0.415 \quad b_{Ge} = 0.84.$$

Observed and calculated intensities are compared in table 2. We have omitted the Debye-Waller factors from our calculations since the 2θ range covered did not exceed 60° . Also a test made for UPd_2Ge_2 has shown that, for $2B = 2.0 \text{ \AA}^{-2}$, no variation of the x parameter occurs which is larger than the fitting errors.

In the cases of UPd_2Si_2 , UPd_2Ge_2 and URh_2Si_2 all observed nuclear reflections obeyed the $h + k + l = 2n$ extinction rule characteristic of a body-centred cell. However, in the case of URh_2Ge_2 , peaks which were comparatively small in intensity with $h + k + l = 2n + 1$ were found on neutron diffraction patterns indicating the symmetry of URh_2Ge_2 to be lower than $I4/mmm$. We have chosen the space group $P4/mmm$ for which the atomic sites are as follows:

1U in 1(a):	0, 0, 0	
1U in 1(d):	$\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$	
2Y in 2(g):	$0, 0, x_1$	$0, 0, \bar{x}_1$
2Y in 2(h):	$\frac{1}{2}, \frac{1}{2}, x_2$	$\frac{1}{2}, \frac{1}{2}, \bar{x}_2$
4Y in 4(i):	$0, \frac{1}{2}, x_3$	$0, \frac{1}{2}, \bar{x}_3$
	$\frac{1}{2}, 0, x_3$	$\frac{1}{2}, 0, \bar{x}_3$

Here Y stands for an 'average' atom with $\langle b \rangle = \frac{1}{2}(b_{Rh} + b_{Ge}) = 0.715 \times 10^{-14}$ m since the final refinement of x_1 , x_2 , x_3 and $\langle b \rangle$ by the least-squares program has uniquely shown that the Ge and Rh ions are distributed at random among the 2(g), 2(h) and 4(i) sites.

4. Magnetic ordering

4.1. UPd_2Si_2 and UPd_2Ge_2

Magnetic reflections on LHT neutron diffraction patterns (see figure 1) appear at scattering angles which correspond to the scattering vectors $\mathbf{H} \pm \mathbf{q}$ (\mathbf{H} is here the scattering vector of a nuclear reflection and \mathbf{q} is the propagation vector of the magnetic

Table 3. The magnetic data of UPd₂Si₂, UPd₂Ge₂ and URh₂Si₂ compounds.

	UPd ₂ Si ₂	UPd ₂ Ge ₂	URh ₂ Si ₂
$T_N(K)$	150 ± 4	140 ± 4	137 ± 4
$\theta_p(K)$ paramagnetic	-62	-81	
Curie temperature			
$\mu_{eff}(\mu_B)$	3.39	3.40	
a/c	0.4078	0.4105	0.3999
$q(c^*) = q(2\pi/c)(\text{\AA}^{-1})$	$q_1 = 0.662 \pm 0.010$ $q_2 = 1.000 \pm 0.010$	0.748 ± 0.010	1.000 ± 0.010
$\lambda = 2\pi/q (\text{\AA})$	$\lambda_1 = 15.18 \pm 0.23$ $\lambda_2 = 10.05 \pm 0.10$	13.69 ± 0.18	10.03 ± 0.10
$A_n(\mu_B)$	$A_1 = 2.05 \pm 0.10$	2.79 ± 0.15	1.96 ± 0.06
4.2 K	$A_2 = 1.32 \pm 0.10$		
$A_n(\mu_B)$	$A_1 = 2.55 \pm 0.10$	2.68 ± 0.10	1.64 ± 0.08
80 K	$A_2 = 0 \pm 0.10$		
$\langle \mu^2 \rangle^{1/2}(\mu_B)$	1.96	1.97	1.96
4.2 K			
$\langle \mu^2 \rangle^{1/2}(\mu_B)$	1.80	1.89	1.64
80 K			

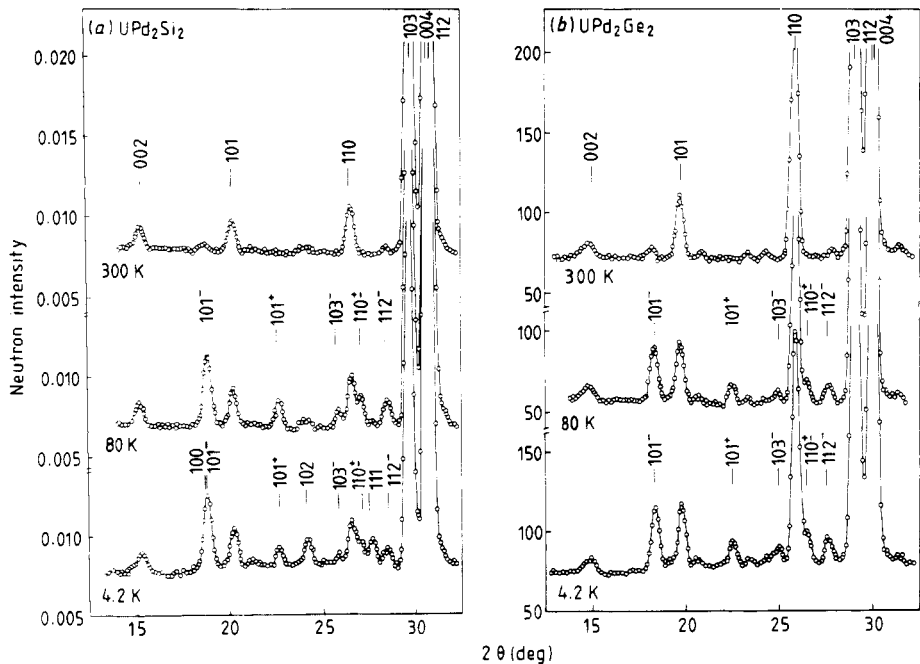


Figure 1. 4.2 K, 80 K and 300 K neutron diffraction patterns of (a) UPd₂Si₂ and (b) UPd₂Ge₂.

structure)†. Vector *q* is paralld to the ordered moments of the U ions, because at all temperatures below the respective Néel points no magnetic peak was observed in the neighbourhood of the reciprocal point *H* = (0,0,0). The direction of the ordered moment is along the tetragonal axis, since the magnetic reflections with (00*l*) indices remain unobserved.

† The symbols *hkl* ± are adopted to denote the positions of magnetic reflections *H* ± *q*.

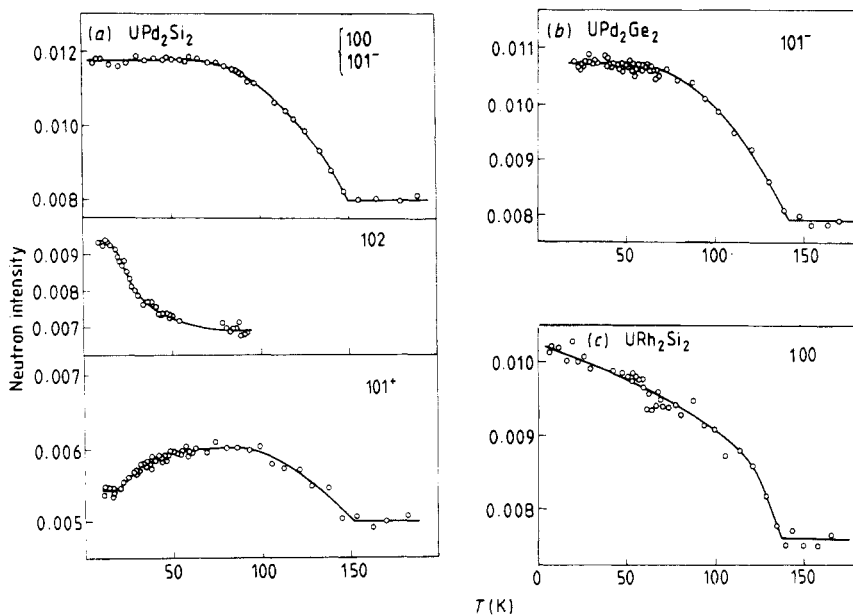


Figure 2. Magnetic peak heights versus temperature for selected reflections in the case of (a) UPd_2Si_2 , (b) UPd_2Ge_2 and (c) URh_2Si_2 .

The positions of the magnetic reflections for both compounds indicate the presence of an incommensurate magnetic structure of a modulated amplitude type, commonly denoted as LSW. The magnetic moment μ of an ion for this type of ordering can be expressed in terms of a Fourier series:

$$\mu_n = \hat{z} \sum_q A_q \cos(2\pi q \cdot R_n + \varphi_q)$$

where \hat{z} denotes a unit vector in the direction of μ , q is a propagation vector of the magnetic structure in the first Brillouin zone, A_q and φ_q are the amplitude and phase angle of the Fourier component of the magnetic moment, and R_n is the position of the n th magnetic ion.

Taking into account that μ is parallel to the c axis and q is parallel to μ , we can write the components of the magnetic moment along coordinate axes chosen in such a way that \hat{z} is parallel to the c axis and x and y are along the a axes of the tetragonal unit cell. Thus:

$$\mu_n^{(x)} = 0 \quad \mu_n^{(y)} = 0 \quad \mu_n^{(z)} = A_1 \cos(2\pi q z_n + \varphi_1).$$

This formula represents the situation found in UPd_2Ge_2 at temperatures between 4.2 K and 149 K and in UPd_2Si_2 between 40 K and 150 K. For the latter compound the 4.2 K neutron pattern contains additional magnetic reflections appearing only at H : their indices are (100), (102) and (111). This implies that the expression for the magnetic moment in this case contains two Fourier components:

$$\mu_n^{(x)} = \mu_n^{(y)} = 0 \quad \mu_n^{(z)} = A_1 \cos(2\pi q_1 z_n + \varphi_1) + A_2 \cos(2\pi q_2 z_n).$$

The amplitudes A_1 and A_2 are temperature dependent.

The values of q_1 and q_2 for UPd_2Si_2 and q for UPd_2Ge_2 were determined by

fitting to the positions of the magnetic reflections. The amplitudes A_i of the Fourier components of the U ion magnetic moment were determined by least-squares fitting. They are listed in table 4. The $5f^2$ form factor was used (Wedgwood 1972). The observed and calculated magnetic intensities are compared in tables 4 and 5. The magnetic structures are displayed in figure 3. In both cases φ_i is assumed to be zero, since only the vectors \mathbf{q}_i and the magnitudes of A_i can be determined from neutron diffraction powder data.

The maximum magnetic moment at the U ion in UPd₂Si₂ amounts to 3.37 ± 0.25 Bohr magnetons (μ_B) at 4.2 K and $2.55 \pm 0.10 \mu_B$ at 80 K. In UPd₂Ge₂ μ_{\max} has a value of $2.79 \pm 0.15 \mu_B$ at 4.2 K and $2.68 \pm 0.10 \mu_B$ at 80 K. The magnitudes of the root-mean-square ordered moment defined as $\langle \mu^2 \rangle^{1/2} = [\frac{1}{2} \sum_q (A_q)^2]^{1/2}$ are listed in table 4.

The Néel points determined from the temperature dependence of the magnetic peak heights are presented in table 3. They show good agreement with the results of magnetic susceptibility measurements (A Zygmunt 1980, private communication).

Table 4. Comparison of the observed and calculated neutron magnetic intensities at 4.2 K. The magnetic contributions to the nuclear peaks were not detected.

<i>hkl</i>	UPd ₂ Si ₂		UPd ₂ Ge ₂		URh ₂ Si ₂	
	$I_{\text{obs}}^m K$	I_{cal}^m $A_1 = 2.05 \mu_B$ $A_2 = 1.32 \mu_B$	$I_{\text{obs}}^m K$	I_{cal}^m $A = 2.79 \mu_B$	$I_{\text{obs}}^m K$	I_{cal}^m $A = 1.96 \mu_B$
100 } 101 ⁻	75.23	70.32	—	—	74.48	69.64
101 ⁺	17.57	17.57	79.41	76.32	—	—
103 ⁻	8.72	9.96	33.83	33.07	—	—
110 [±]	18.98	15.97	20.04	20.23	—	—
112 ⁻	11.84	12.56	29.11	31.04	—	—
102	21.93	21.80	25.66	25.60	51.99	48.67
111	23.55	23.68	—	—	43.44	50.39

Table 5. Comparison of the observed and calculated neutron magnetic intensities at 80 K. The magnetic contributions to the nuclear peaks were not detected.

<i>hkl</i>	UPd ₂ Si ₂		UPd ₂ Ge ₂		URh ₂ Si ₂	
	$I_{\text{obs}}^m K$	I_{cal}^m $A_1 = 2.55 \mu_B$ $A_2 = 0$	$I_{\text{obs}}^m K$	I_{cal}^m $A = 2.68 \mu_B$	$I_{\text{obs}}^m K$	I_{cal}^m $A = 1.64 \mu_B$
100	—	—	—	—	52.11	48.75
101 ⁻	65.42	58.98	77.99	70.44	—	—
101 ⁺	23.43	27.18	25.41	30.51	—	—
103 ⁻	10.67	15.41	19.79	18.67	—	—
110 [±]	26.46	24.71	32.04	28.65	—	—
112 ⁻	19.80	19.44	23.95	23.62	—	—
102	—	—	—	—	30.53	34.08
111	—	—	—	—	33.85	35.28

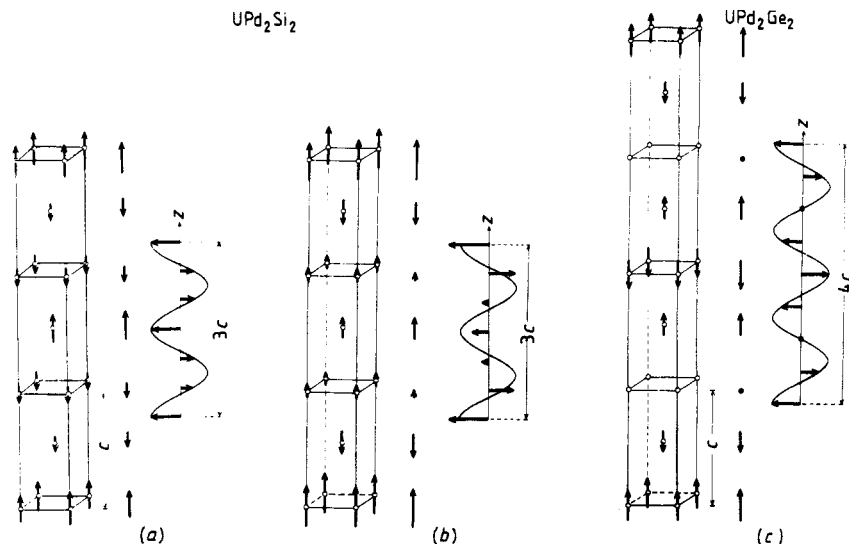


Figure 3. Schematic representation of the magnetic structures of (a) UPd_2Si_2 at 4.2 K, (b) UPd_2Si_2 at 80 K, (c) UPd_2Ge_2 at temperatures below T_N . The phase angle ϕ_i was assumed to be zero.

The temperature dependence of the magnetic reflections corresponds to the temperature dependence of $A_i^2(T)$. In UPd_2Si_2 the moment amplitudes $A_1(T)$ and $A_2(T)$ exhibit a different variation with temperature: at 40 ± 5 K the ordered moment amplitude A_2 becomes zero. Consequently, above this temperature we observe only magnetic peaks with $\mathbf{H} \pm \mathbf{q}_1$.

Since the magnetic peaks $\mathbf{H} \pm \mathbf{q}_1$ and $\mathbf{H} \pm \mathbf{q}_2$ appeared at different positions on our 4.2 K neutron pattern we were able to trace the temperature behaviour of $A_1(T)$ and $A_2(T)$ independently and then calculate the intensity of the overlapping reflections with indices (100) and (101) $^-$. This is shown in figure 4, whereas the measured dependence is displayed in figure 2(a).

4.2 URh_2Si_2

Both the 4.2 K and 80 K neutron diffraction patterns indicate an antiferromagnetic order, since the indices of the magnetic reflections obey the extinction rule $h + k + l = 2n + 1$. Neither magnetic reflections with indices (00l) nor magnetic contributions to nuclear peaks were observed, so that it may be concluded that the magnetic and chemical unit cells coincide, the magnetic moment alignment being along the tetragonal axis. The propagation vector of such a magnetic structure \mathbf{q} is (0, 0, 1) c^* . The magnetic structure can be simply described as a succession of ferromagnetic sheets in the c direction with the sequence $+ -$. Magnetic parameters of URh_2Si_2 are given in table 3.

4.3. URh_2Ge_2

Despite the results of susceptibility measurements, our neutron diffraction study shows that URh_2Ge_2 is in a non-ordered state at 4.2 K in zero magnetic field. Neither separate magnetic reflections or contributions to nuclear peaks were detected.

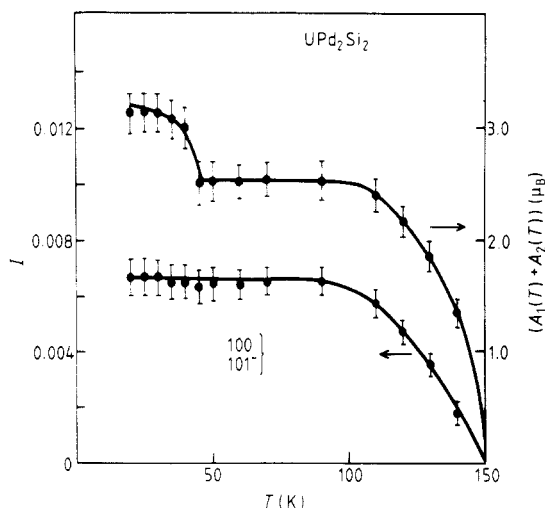


Figure 4. The calculated sum $A_1(T) + A_2(T)$ for UPd_2Si_2 from observed temperature dependences of the 102 and 101^+ magnetic reflections (the upper curve) and the calculated intensity temperature dependence of overlapping 100 and 101^- reflections (the lower curve).

5. Discussion

In general the oscillatory character of the magnetic ordering in alloy systems is considered due to the oscillatory nature of the magnetic interactions via conduction electrons. Such a mechanism is described by the RKKY theory (Ruderman and Kittel 1954, Kasuya 1956, Yosida 1957).

This theory was adopted by Grunzweig-Genossar *et al* (1968) and Grunzweig-Genossar and Cahn (1973) to interpret the magnetic properties of UX compounds. We have attempted to check whether the magnetic structures with $0 < q < 1$ can be stable in the tetragonal body-centred crystal lattice, assuming only the RKKY model of interactions. We have taken into account the following arguments.

(i) In all the alloys studied the shortest U–U distances are quite large: they amount to about 4 Å between uraniums in the basal plane and to about 5.8 Å between the U ion in the centre of the unit cell and a U ion at its corner. The latter are separated by three layers of non-magnetic ions. For these reasons the occurrence of direct or superexchange interactions between U ions seems very improbable.

(ii) Magnetic measurements performed on the isostructural compound ThPd_2Si_2 shows that Pd ions do not carry a magnetic moment (A Zygmunt 1980, private communication). This justifies our assumption that magnetic interactions take place only among U ions.

(iii) UPd_2Si_2 exhibits relatively high electrical conductivity (A Zygmunt 1980, private communication).

(iv) Our neutron diffraction data show that in UPd_2Si_2 and UPd_2Ge_2 the magnetic ordering has an oscillatory character.

(v) The magnetic moment is localised on the U ion and it is adopted in discussions on the magnetic properties of uranium intermetallics (see, for example, Freeman *et al* 1979, de Novion 1979).

The RKKY theory shows that the energy E of a spin system with a screw-like ordering is given by

$$E = -NS^2J(\mathbf{q}) \sim -(J(\mathbf{q}) - J(0)) \sim -F(\mathbf{q})$$

where N is the total number of magnetic ions, S is the magnitude of the spin and $J(\mathbf{q})$ represents the Fourier transform of the exchange integral $J(\mathbf{R}_j - \mathbf{R}_i)$ between two spins; the function $F(\mathbf{q})$ is described in detail by Yosida and Watabe (1962). Assuming that J is a constant of motion for electrons in the 5f state, S can be replaced by $(g - 1)J$, where g is the Landé factor.

The other basic assumption is that the Fermi surface is spherical. This means that the length of the Fermi vector $k_f = (\pi/a)(6za/\pi c)^{1/3}$ is dependent on the number z of free electrons per magnetic ion.

The fact that a screw-type ordering is stable means that the function $F(\mathbf{q})$ exhibits a maximum for a non-zero value of q_0 .

The function $F(\mathbf{q})$ was computed for $\mathbf{q} = (0, 0, q)$ and $\mathbf{q} = (q, 0, 0)$. In constructing the reciprocal space, x-ray determined ratios a/c which amount to 0.4078, 0.4105 and

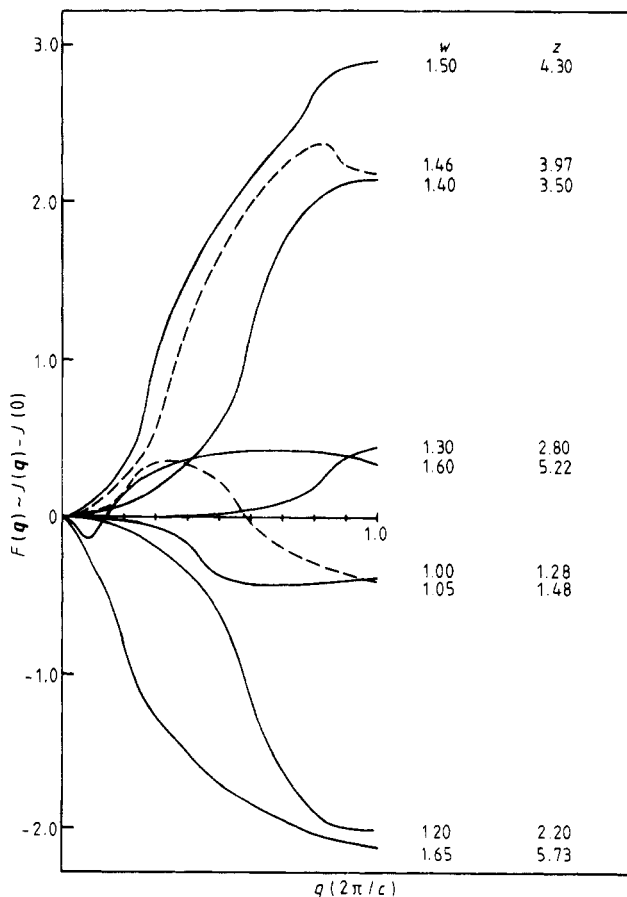


Figure 5. The function $F(\mathbf{q})$ for different values of z in the case of UPd_2Ge_2 . $a/c = 0.4105$, $\mathbf{q} = (0, 0, q)$. Values of w are given against each curve where w is given by the equation $2k_f = w(2\pi/a)$.

0.3999 for UPd_2Si_2 , UPd_2Ge_2 and URh_2Si_2 respectively were used. The summation was carried out over reciprocal lattice points up to $H = 7.5(2\pi/a) = 18.7(2\pi/c)$.

Computations show that for $\mathbf{q} = (q, 0, 0)$ the function $F(\mathbf{q})$ has a maximum only when $q = 0$ or $q = 1$. This means that only a ferromagnetic or a collinear antiferromagnetic structure is stable.

Different behaviour was found for $\mathbf{q} = (0, 0, q)$. Two narrow regions of z (or $2k_f$) are observed (figure 5), for which $F(\mathbf{q})$ exhibits maxima. For example, in the case of UPd_2Ge_2 ($a/c = 0.4105$) they are present when $2k_f = 1.46(2\pi/a)$ (i.e. for $z = 3.97$) and when $2k_f = 1.05(2\pi/a)$ ($z = 1.48$).

The values of q_0 at which the maxima are observed are z dependent. This is shown in figure 6. Only previously mentioned regions are specified. For example, for UPd_2Ge_2 the observed value of the magnetic structure vector \mathbf{q}_0 is 0.748 ± 0.010 (in c^*). It corresponds to $z = 3.86$ free electrons per U ion. For the same value of z , the theoretical value of \mathbf{q}_0 in the case of UPd_2Si_2 is 0.69 c^* , i.e. very close to the observed value of the magnetic structure vector of $(0.662 \pm 0.010)c^*$. However, for URh_2Si_2 our computations indicate that for $z = 3.86$ the maximum of $F(\mathbf{q})$ at $q = 1$, i.e. a collinear antiferromagnetic ordering with a $+$ $-$ sequence of ferromagnetic sheets should be stable, in accordance with experiment.

Considering the fact that in these computations a spherical Fermi surface and only isotropic RKKY type interactions have been assumed, the above quantitative relations might be quite coincidental; however, in general they indicate that the interactions via conduction electrons are dominant in UMe_2X_2 intermetallics. Of course, more realistic considerations could be carried out when detailed data of the Fermi surface shape and bandstructure in UMe_2X_2 intermetallics are available.

The root mean square of the magnetic moment on the U ion at 4.2 K was determined to be the same within the fitting error in all three former compounds amounting to $1.96 \pm 0.10 \mu_B$. This and only small differences among the observed Néel points incline us to conclude that the magnetic interactions are of the same kind in all the three compounds investigated. Simple theoretical considerations in which only RKKY interactions were allowed for in the spin Hamiltonian suggest that the observed

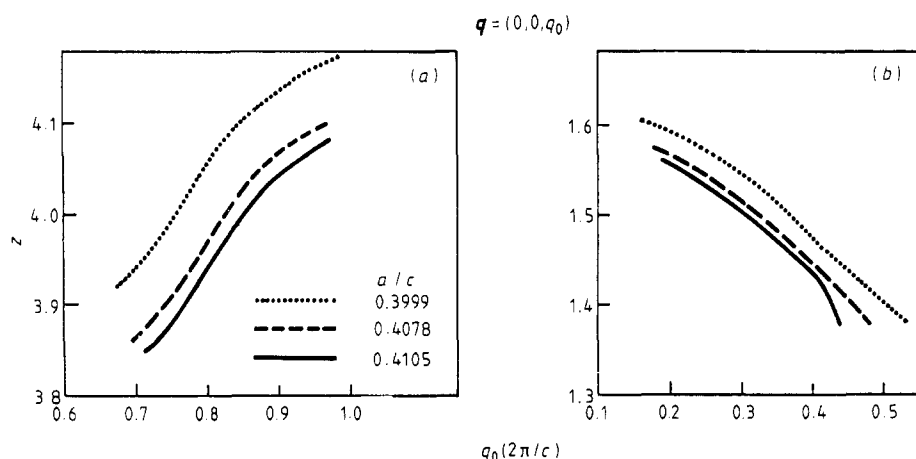


Figure 6. The curves representing the dependence of vector \mathbf{q}_0 on z calculated for UPd_2Si_2 (broken curve), UPd_2Ge_2 (full curve) and URh_2Si_2 (dotted curve). The function $F(\mathbf{q})$ exhibits a maximum at \mathbf{q}_0 for which a screw-like structure is stable. (a) and (b) represent the regions of z values for which the screw-like magnetic structures are stable.

changes in the magnetic ordering schemes and their stability are due to isotropic interactions via conduction electrons.

The RKKY interactions favour an incommensurate moment alignment. On the other hand, the crystal field effect is usually commensurate with the crystal structure. It becomes stronger as the temperature is lowered. This effect is probably responsible for the appearance of a commensurate magnetisation wave with $\mathbf{q} = (0, 0, 1)c^*$ observed in UPd_2Si_2 below 40 K.

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