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MAGNETISM IN URANIUM COMPOUNDS OF Th_3P_4 CRYSTAL STRUCTURE

Cz. Oleksy, J. Przystawa
Instytut Fizyki Teoretycznej Uniwersytetu Wrocławskiego,
Pl. Maksa Borna 9, 50-204 Wrocław, Poland

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The present paper discusses the magnetism of the U_3X_4 compounds (X = P, As, Sb, Bi) in terms of an Heisenberg–type model introduced in 1987 by the authors. It contains competing single–ion and exchange isotropic and anisotropic interactions of comparable magnitude. The model is derived using a symmetry analysis of the structures and the Landau theory of phase transitions. The results of the mean–field calculations are presented, describing the spontaneous magnetisation, temperature dependence phase diagrams and behaviour of the system in the external magnetic field. Also the elementary excitations are discussed. The model provides a good description of the magnetic structures and their thermodynamic behaviour.

Key words: Heisenberg model, magnetic phase transitions, spin waves.

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I. INTRODUCTION

Physical properties of magnetically ordered Uranium compounds with the Th_3P_4-crystal structure have received attention since the pioneering work of Trzebiatowski and Troć [1] who discovered a ferromagnetic transition in U_3P_4 at about 140 K. Subsequently, ferromagnetic properties were revealed in other compounds of this family, i.e. with X=As, Sb, Bi [2–4] as well as with X =Se and Te [5, 6]. The nature of the magnetic phases of these compounds posed a more difficult problem. First neutron diffraction experiments on powder samples of U_3P_4 suggested a collinear ferromagnetic structure [7, 8]. A similar conclusion was reached regarding U_3As_4 [9] and U_3Sb_4 [10]. However, preliminary theoretical analysis of the phase transition lead to the conclusion that one should expect a non-collinear multi-axial structures rather than a simple ferromagnetism [11, 12].

A long controversy as to this question was eventually resolved by Burlet et al. [13] using polarized neutron spectroscopy of monocrystal samples of U_3P_4 and U_3As_4: the magnetic structures in both materials were found to be non-collinear three-axial ferromagnets. On the other hand, similar recent experiments on U_3Sb_4 and U_3Bi_4 decided in favour of collinear structures [14]. Samples of U_3Te_4 and U_3Se_4 proved to be more difficult to handle and the question of what are the magnetic structures of these materials remains unresolved.

Although all U_3X_4 materials are cubic crystals of T_d^6 ($I\bar{4}3d$) cubic space group, they turn out to be highly anisotropic [15–19] and this was confirmed both by magnetic [15–19] and by electrical measurements [20].

Giant magnetic anisotropy has been observed in a number of experiments conducted in high magnetic fields [21–26]. The semimetallic nature of these solid compounds is also well established [20, 25, 27–29].

Theoretical attempts to explain physical properties of the magnetically ordered states of the U_3X_4 compounds have followed two paths: (1) In view of their semimetallic nature, Adachi and Imoto [30] exploited the Ruderman–Kittel–Kasuya–Yosida (RKKY) theory [31–33]. Unfortunately, perhaps as a result of a profound disagreement with the experimental data (in the case of U_3X_4 compounds), this approach was abandoned for the next two decades. It was only recently that the itinerant electron model was used to calculate the magnetic structure of U_3P_4 by Sandratskii and Kübler [34] and they were able to discuss non-collinear ferromagnetism within the local approximation to spin–density functional theory. However, the angle of non-collinearity they obtained was much smaller than the one observed in the experiment of Burlet et al [13].

An alternative approach to explain magnetic phenomena in the U_3X_4 compounds is to discuss them within a framework of a localised spin Heisenberg–type model. The first such model was proposed by Przystawa and Praveczki [35]. Their model, however, reduced the cubic symmetry of the Hamiltonian to a rhombohedral one and, on top of that, they decoupled the single-ion crystal field anisotropy in the molecular field approximation like the exchange interaction. A somewhat better model was used by Walasek et al. [36] to analyse the phase transition in U_3X_4. Neglecting the exchange interaction anisotropy, they preserved the cubic symmetry of the Hamiltonian and also the single-ion anisotropy was treated exactly. However, they applied high temperature perturbation theory, where the effective exchange interaction was treated as a perturbation. This limited the validity of their approach to a region not too distant from the transition temperature and could not account for giant anisotropy which was detected in experiments. The same approach was used by Pawlikowski et al. [37] who supplemented the Przystawa and Praveczki Hamiltonian [35] by higher order crystal–field terms.

A significant improvement of the theory was achieved with a model that was first introduced in ref. [25]. This new model, by comparison with the Hamiltonian used by Walasek et al [36], contained a new type of exchange anisotropy compatible with the overall cubic symmetry of the crystal. Subsequent calculations based on this Hamiltonian were able to provide an explanation of a number of properties, ground state phase diagrams [25], phase transitions [38], behaviour in the high magnetic fields [25, 38] and spin–wave excitations [39].

This paper is devoted to the presentation of this model, its derivation and application to the behaviour of the U_3X_4 systems in the magnetically ordered states.

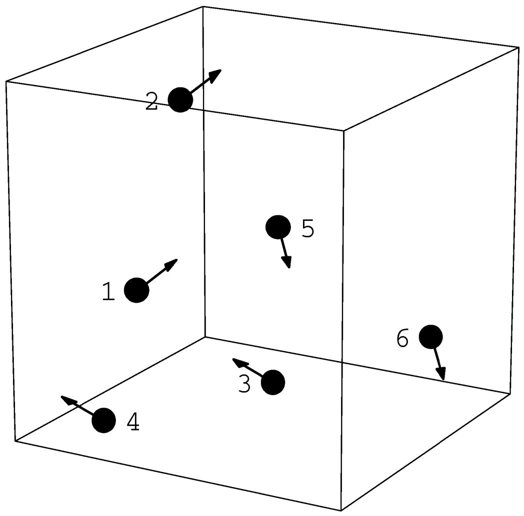

Fig. 1. The unit cell of the magnetically ordered U_3P_4.

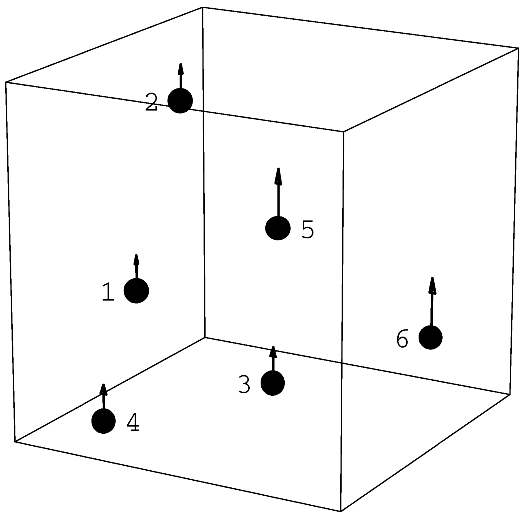

Fig. 2. The unit cell of the magnetically ordered U_3Sb_4.

The paper is organized as follows: In section 2, we present the symmetry analysis of possible magnetic structures in U_3X_4 crystals limited to $\vec{k}=0$ case, i.e. pertinent to magnetic ordering with a ferromagnetic component. In section 3, the Landau theory of symmetry changes that gives two magnetic orderings corresponding to those found in U_3X_4 with X = P, As, Sb and Bi is presented. In section 4, the model Heisenberg–type Hamiltonian is constructed, which contains the crystal–field three-axial anisotropy and a new cubic exchange–type anisotropy. In section 5 the mean–field phase diagram at T=0 is described. Section 6 presents the behaviour of the system in external magnetic fields at T=0. In section 7, we discuss temperature–dependent phase diagrams, obtained in the mean–field approximation. Section 8 discusses the elementary excitations (spin–waves) in both non-collinear and collinear phases. Section 9 contains some conclusions.

II. SYMMETRY ANALYSIS OF THE SPIN SYSTEM OF U_3X_4

In what follows we use the term "spin" to denote localised magnetic moments of the Uranium ions as only their transformation properties with respect to the space group symmetry operations will be important.

The crystal structure of all U_3X_4 compounds are shown in Figs. 1 and 2, where the positions of the magnetic ions in the unit cell of the crystal are depicted.

The magnetic ions are situated at the following special positions in the crystallographic unit cell:

$$\vec{r}_1=(\tfrac{3}{8},0,\tfrac{1}{4}),\ \vec{r}_2=(\tfrac{1}{8},0,\tfrac{3}{4}),\ \vec{r}_3=(\tfrac{1}{4},\tfrac{3}{8},0),$$

$$\vec{r}_4=(\tfrac{3}{4},\tfrac{1}{8},0),\ \vec{r}_5=(0,\tfrac{1}{4},\tfrac{3}{8}),\ \vec{r}_6=(0,\tfrac{3}{4},\tfrac{1}{8}).$$

They are transformed among themselves by the symmetry operations of the bcc cubic space group I$\bar{4}$3d-T_d^6 (see, e.g., Refs. [40]). Following Refs. [41–44], we perform a symmetry analysis of a general spin structure of the formula

$$\vec{M}(\vec{r})=\sum_{i=1}^{6}\vec{S}_i\delta(\vec{r}-\vec{r}_i) \tag{1}$$

where $\vec{S}_i$ denotes an arbitrary spin vector at the site $\vec{r}_i$. As all magnetic structures of U_3X_4 compounds are ferro (ferri) magnetic it is sufficient to consider only the spin in the unit cell, i.e. we take into account only structures with $\vec{k}=0$, i.e. at the Γ-point of the Brillouin Zone. If we apply all symmetry operations of the group I$\bar{4}$3d to the spin structure (1), we find 18 equivalent structures that form a basis of an 18-dimensional representation $D(\bar{G})$ of the group I$\bar{4}$3d-T_d^6. The group has 5 irreducible representations at Γ (see table 1).

The symmetry group operations are given in the standard notation. With standard methods (see, e.g., [41]), the reducible representation $D(\bar{G})$ can be decomposed into

	E	C_3	C_{2x}	C_{4z}	C_{2a}
τ^1	1	1	1	1	1
τ^2	1	1	1	-1	-1
τ^3	2	$\begin{pmatrix} \epsilon^2 & 0 \\ 0 & \epsilon \end{pmatrix}$	$\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$	$\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$
τ^4	3	$\begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}$	$\begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$	$\begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}$	$\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$
			$\tau^5 = \tau^4 \otimes \tau^2$		
			with $\epsilon = \exp(i2\pi/3)$		

Table 1. Irreducible representations of the group I$\bar{4}$3d at the Γ-point.

$$D(\bar{G}) = \tau^1 \oplus \tau^3 \oplus \tau^4 \oplus \tau^4 \oplus \tau^5 \oplus \tau^5 \oplus \tau^5. \tag{2}$$

What structures can be described by these irreducible representations? This question was answered in Refs. [42, 43]: only τ^5 can describe a ferromagnetic system, i.e. one with a resulting magnetic moment different from zero. The other lead to structures with net magnetic moment equal to zero, i.e. some sort of antiferromagnetic arrangement. Thus we must discuss only structures compatible with the τ^5 irreducible representation. However, as we see from Eq. (2) τ^5 appears three times in the decomposition of D and thus couplings between the different parameters of the same symmetry may be necessary.

Using the projection-operator technique, the basis functions of different τ^5 representations can be constructed. The following 3 independent bases can be chosen [44]:

$$\sigma_1 = \frac{1}{2}(S_1^x + S_2^x),$$

$$\sigma_2 = \frac{1}{2}(S_3^y + S_4^y),$$

$$\sigma_3 = \frac{1}{2}(S_5^z + S_6^z),$$

$$\rho_1 = \frac{1}{4}(S_3^x + S_4^x + S_5^x + S_6^x),$$

$$\rho_2 = \frac{1}{4}(S_1^y + S_2^y + S_5^y + S_6^y),$$

$$\rho_3 = \frac{1}{4}(S_1^z + S_2^z + S_3^z + S_4^z),$$

$$\gamma_1 = \frac{1}{4}(-S_3^z + S_4^z + S_5^y - S_6^y),$$

$$\gamma_2 = \frac{1}{4}(S_1^z - S_2^z - S_5^x + S_6^x),$$

$$\gamma_3 = \frac{1}{4}(-S_1^y + S_2^y + S_3^x - S_4^x), \tag{3}$$

where S_i^α denotes here the α-th component of a unit spin vector at the site $\vec{r}_i$. The magnetic structure proposed by Przystawa [11] for U_3P_4 was

$$\vec{S}_1 = \begin{pmatrix} u \\ v \\ w \end{pmatrix}, \quad \vec{S}_2 = \begin{pmatrix} u \\ w \\ v \end{pmatrix}, \quad \vec{S}_3 = \begin{pmatrix} w \\ u \\ v \end{pmatrix},$$
$$\vec{S}_4 = \begin{pmatrix} v \\ u \\ w \end{pmatrix}, \quad \vec{S}_5 = \begin{pmatrix} v \\ w \\ u \end{pmatrix}, \quad \vec{S}_6 = \begin{pmatrix} w \\ v \\ u \end{pmatrix}. \tag{4}$$

In terms of the basis functions (3), this structure can be written as follows

$$\vec{m}(\vec{r}) = u(\sigma_1 + \sigma_2 + \sigma_3) + (v + w)(\rho_1 + \rho_2 + \rho_3)$$
$$+(w - v)(\gamma_1 + \gamma_2 + \gamma_3),$$

and the actual structure of U_3P_4 and U_3As_4 [12, 13] is obtained by putting $v = w$ (see fig. 1)

$$\vec{m}(\vec{r}) = u\sum_{i=1}^{3}\sigma_i + 2v\sum_{i=1}^{3}\rho_i\ , \tag{5}$$

Such a magnetic structure is invariant with respect to the symmetry operation of the magnetic space group R3c′ [11] and is entirely described by the 3-dimensional irreducible representation of I$\bar{4}$3d, i.e. τ^5.

III. THE LANDAU THEORY OF SYMMETRY CHANGES IN U_3X_4

A general discussion of usefulness and limitations of the Landau theory of symmetry changes can be found in ref. [45]. The classical Landau rules [46] require that the order parameters belong to an irreducible representation of the paramagnetic space group for the transition to be of the second order. In our case, the function (5) (which plays the role of the order parameter) involves the irreducible representation τ^5 at least twice. This, however, does not exclude the continuity of the phase transition (see, e.g., ref. [45]).

Preliminary Landau analysis of the transition in U_3X_4 was done by Przystawa [11], Przystawa and Cracknell [43] and in the most complete way by Oleksy [44].

Using the basis functions (3), we construct the Landau free energy invariants with respect to the symmetry operations of the group I$\bar{4}$3d. The requirement that the Hamiltonian have time–reversal invariance excludes from the Hamiltonian any odd couplings and only bilinear and quartic terms will appear, viz.

$$\Phi(\sigma,\rho,\gamma) = \Phi_2(\sigma,\rho,\gamma) + \Phi_4(\sigma,\rho,\gamma) \tag{6}$$

with

$$\begin{aligned}\Phi_2(\sigma,\rho,\gamma) = {} & a_1 I(\sigma^2) + a_2 I(\rho^2) + a_3 I(\gamma^2) \\ & + 2kI(\sigma\rho) + 2k_1 I(\sigma\gamma) + 2k_2 I(\rho\gamma),\end{aligned} \tag{7}$$

where

$$I(\sigma\rho) \equiv \sum_{i=1}^{3} \sigma_i \rho_i, \quad \text{etc.}$$

We shall not write down the quartic terms, as they are quite lengthy and their explicit form will not be needed in this presentation. The full potential (6) has been discussed in ref. [44]. The results are not qualitatively different from the analysis of a simplified form, in which the two couplings, k_1 and k_2 are neglected. It is always a subtle question of how to justify such a simplification. On the one hand, the γ-part of the symmetry–allowed order parameter (3) does not contribute to the actual structure of U_3P_4 (5). On the other hand, in what follows, we shall proceed along the lines of the Landau School [47–49], to select various interactions contributing to (7) based upon their relative magnitude. To do so one should rewrite the invariants (7) in terms of S_i^α rather than σ, ρ and γ and divide the contributing terms into "exchange forces" and "relativistic forces". In such a way we simplify (6) to

$$\begin{aligned}\Phi(\sigma,\rho) = {} & a_1 I(\sigma^2) + a_2 I(\rho^2) \\ & + 2kI(\sigma\rho) + \Phi_4(\sigma,\rho).\end{aligned} \tag{8}$$

There are only two structures that minimize the free energy

<u>Solution 1: The C-structure</u> (fig. 1)

$$\begin{gathered}\vec{S}_1 = \vec{S}_2 = \begin{pmatrix}\sigma\\ \rho\\ \rho\end{pmatrix}, \quad \vec{S}_3 = \vec{S}_4 = \begin{pmatrix}\rho\\ \sigma\\ \rho\end{pmatrix}, \\ \vec{S}_5 = \vec{S}_6 = \begin{pmatrix}\rho\\ \rho\\ \sigma\end{pmatrix},\end{gathered} \tag{9}$$

i.e. a three-axial ferromagnetic structure with a magnetic moment along the [111] direction, i.e.

$$\vec{M} = \sum_{i=1}^{6} \vec{S}_i = 2\begin{pmatrix}\sigma+2\rho\\ \sigma+2\rho\\ \sigma+2\rho\end{pmatrix}.$$

This is the structure determined in [13] for U_3P_4 and U_3As_4. The magnetic symmetry of this structure is R3c′ [11].

<u>Solution 2: The L-structure</u> (fig. 2)

$$\begin{aligned}&\vec{S}_1 = \vec{S}_2 = \vec{S}_3 = \vec{S}_4 = \begin{pmatrix}0\\ 0\\ \rho'\end{pmatrix}, \\ &\vec{S}_5 = \vec{S}_6 = \begin{pmatrix}0\\ 0\\ \sigma'\end{pmatrix},\end{aligned} \tag{10}$$

i.e. a collinear ferrimagnetic structure with a magnetic moment along the [001] direction, i.e.

$$\vec{M} = \sum_{i=1}^{6} \vec{S}_i = 2\begin{pmatrix}0\\ 0\\ \sigma'+2\rho'\end{pmatrix}.$$

This is the structure reported in [14] for U_3Sb_4 and U_3Bi_4. The magnetic symmetry of this structure is I$\bar{4}$2'd' [11].

It is interesting to note [44] that the minimization of the full quartic potential (6) only slightly changes the form of the two above solutions. <u>Solution 1</u> becomes 6-axial (4) with

$$u = \sigma, \;\; v = \rho - \gamma \;\; \text{and} \;\; w = \rho + \gamma,$$

while solution 2 becomes slightly antiferromagnetic in the plane perpendicular to [001]

$$\vec{S}_1 = \begin{pmatrix} 0 \\ -\gamma' \\ \rho' \end{pmatrix}, \quad \vec{S}_2 = \begin{pmatrix} 0 \\ \gamma' \\ \rho' \end{pmatrix}, \quad \vec{S}_3 = \begin{pmatrix} \gamma' \\ 0 \\ \rho' \end{pmatrix},$$

$$\vec{S}_4 = \begin{pmatrix} -\gamma' \\ 0 \\ \rho' \end{pmatrix}, \quad \vec{S}_5 = \vec{S}_6 = \begin{pmatrix} 0 \\ 0 \\ \sigma' \end{pmatrix} \tag{11}$$

but the net magnetic moment remains along the [001] direction and the symmetry group of the structure does not change.

IV. THE MODEL HAMILTONIAN

In looking for a suitable model Hamiltonian to describe the magnetically ordered state of our materials we follow the procedure proposed by Dzyaloshinsky et al. [47–49], i.e. we shall rewrite the potential (8) in terms of S_i^α, representing the α-th component of the spin in the i-th sublattice. As we are looking for a bilinear spin–Hamiltonian, we shall consider only bilinear contributions to $\Phi(\sigma,\rho)$. In such a way we obtain numerous bilinear contributions in terms of the spin operators. These terms correspond to various physical "spin–spin" interactions, which are allowed by the overall symmetry of the Hamiltonian. Of course, we must select these terms by their relative magnitude. Following Dzyaloshinsky we divide these contributing forces into 'exchange forces' and 'relativistic forces'. We retain only the nearest–neighbour exchange interactions; we also keep the crystal–field single–ion term and the nearest–neighbour "exchange-type" anisotropy. In such a way the potential (8) may be written as

$$\begin{aligned}\Phi(\vec{S}_1,\ldots,\vec{S}_6) &= \tau\sum_{\alpha=1}^{3}\sum_{i=1}^{6}(\vec{S}_i^\alpha)^2 \\ &+D[(S_1^x)^2+(S_2^x)^2+(S_3^y)^2+(S_4^y)^2+(S_5^z)^2+(S_6^z)^2] \\ &+J[(\vec{S}_1+\vec{S}_2)(\vec{S}_5+\vec{S}_6)+(\vec{S}_1+\vec{S}_2)(\vec{S}_3+\vec{S}_4)+(\vec{S}_3+\vec{S}_4)(\vec{S}_5+\vec{S}_6)] \\ &+K[(S_1^y+S_2^y)(S_5^y+S_6^y)+(S_1^z+S_2^z)(S_3^z+S_4^z)+(S_3^x+S_4^x)(S_5^x+S_6^x)]\end{aligned} \tag{12}$$

with

$$\tau=\frac{1}{16}a_1,\quad D=\frac{1}{16}(4a_1-a_2),\quad J=\frac{1}{4}k,\quad K=\frac{1}{8}(a_2-2k).$$

It is perhaps worth mentioning that the complete Dzyaloshinsky–Moriya term [50, 51], which was suggested by Sandratskii and Kübler [34] as a possible source of the non-collinearity of the magnetic ordering in U_3P_4 is not detected by this consideration. Among the relativistic interactions neglected is a 'broken' Dzyaloshinsky–Moriya term of the following form

$$\alpha[(\vec{S}_1\times\vec{S}_2)_x+(\vec{S}_3\times\vec{S}_4)_y+(\vec{S}_5\times\vec{S}_6)_z]. \tag{13}$$

Thus, basing on (12) we propose the following model Hamiltonian to describe the magnetic properties of the U_3P_4 systems:

$$\begin{aligned}\mathcal{H} = &-\frac{1}{2}\sum_{i,j}\sum_{g_i,g_j}\sum_{\alpha}J_{g_ig_j}^{\alpha\alpha}S_{g_i}^\alpha S_{g_j}^\alpha \\ &-\sum_{i,g_i,\alpha}H^\alpha S_{g_i}^\alpha+\mathcal{H}_{CF}\end{aligned} \tag{14}$$

where $S_{g_i}^\alpha$ — is the αth component of the spin operator at the g-th site in the sublattice ($i=1,2,\ldots,6$), $J_{g_ig_j}^{\alpha\alpha}$ — is the anisotropic interaction tensor, H^α — is the αth component of the external magnetic field, $\mathcal{H}_{CF}$ — is the crystal field Hamiltonian, viz.

$$\mathcal{H}_{CF}=-D\sum_{i,g_i}\left(S_{g_i}^{\mu(i)}\right)^2 \tag{15}$$

where

$$\mu(i)=\begin{cases} x & \text{for } i=1,2, \\ y & \text{for } i=3,4, \\ z & \text{for } i=5,6. \end{cases}$$

The exchange interaction tensor can be specified as

$$\hat{J}_{ii} = 0 \quad \text{for} \quad i = 1, 2, \ldots, 6,$$

$$\hat{J}_{12} = \hat{J}_{34} = \hat{J}_{56} = 0,$$

$$\hat{J}_{13} = \hat{J}_{14} = \hat{J}_{23} = \hat{J}_{24} = \begin{pmatrix} J & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & K \end{pmatrix},$$

$$\hat{J}_{15} = \hat{J}_{16} = \hat{J}_{25} = \hat{J}_{26} = \begin{pmatrix} J & 0 & 0 \\ 0 & K & 0 \\ 0 & 0 & J \end{pmatrix},$$

$$\hat{J}_{35} = \hat{J}_{36} = \hat{J}_{45} = \hat{J}_{46} = \begin{pmatrix} K & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J \end{pmatrix}.$$

Thus our Hamiltonian contains only the nearest–neighbour exchange interactions. However, a novel feature is the occurrence of a new anisotropic exchange interaction, which should not be confused with the one in ref. [35] or ref. [37]: it is a bilinear exchange type anisotropy compatible with the cubic symmetry of the crystal. It is generally believed that cubic symmetry excludes any bilinear anisotropy of the exchange type. The Th_3P_4 structure, however, due to the special positions occupied by magnetic ions, breaks this rule and allows such an anisotropy to be present.

V. MEAN–FIELD THEORY OF SPONTANEOUS MAGNETISATION

We shall describe the thermodynamic properties of the U_3X_4 magnetics in the Molecular Field Approximation (MFA).

In the MFA, our model Hamiltonian would be replaced by the following effective Hamiltonian

$$\tilde{\mathcal{H}} = \tilde{\mathcal{H}}_{CF} + \mathcal{H}_{ex} \tag{16}$$

where

$$\mathcal{H}_{ex} = -\sum_{i,g_i,\alpha} n_i^\alpha S_{g_i}^\alpha. \tag{17}$$

Here $\tilde{\mathcal{H}}_{CF}$ differs from Eq. (15) in the replacement of the crystal–field anisotropy constant D by $d = \frac{D}{zJ}$ where z is the number of nearest neighbours in g_i.

The effective field $\vec{n}_i$ acting on the magnetic ions on the sublattice i is determined by the minimization of the trial free energy [36, 38]

$$\mathcal{F} = -\frac{1}{b}\ln Tr\{\exp[-b\tilde{\mathcal{H}}]\} + \frac{Tr[\exp(-b\tilde{\mathcal{H}})(\frac{\mathcal{H}}{zJ} - \tilde{\mathcal{H}})]}{Tr\exp(-b\tilde{\mathcal{H}})} \tag{18}$$

with $b = \frac{zJ}{k_B T}$.

This requirement yields the self-consistent equations

$$n_i^\alpha = \sum_j J_{ij}^{\alpha\alpha} m_j^\alpha, \tag{19}$$

$$m_i^\alpha = -\frac{\partial \mathcal{F}_i}{\partial n_i^\alpha}, \tag{20}$$

where

$$J_{ij}^{\alpha\alpha} = \frac{1}{zJ}\sum_{\delta_j} J_{g_i,g_i+\delta_j}^{\alpha\alpha}, \tag{21}$$

$$\mathcal{F}_i = -\frac{1}{b}\ln Tr\{\exp[-b\mathcal{H}_i]\}, \tag{22}$$

$$\mathcal{H}_i = -dQ_{g_i} - \sum_\alpha n_i^\alpha S_{g_i}^\alpha. \tag{23}$$

The summation in Eq. (21) runs over the z nearest neighbours of g_i on the sublattice j.

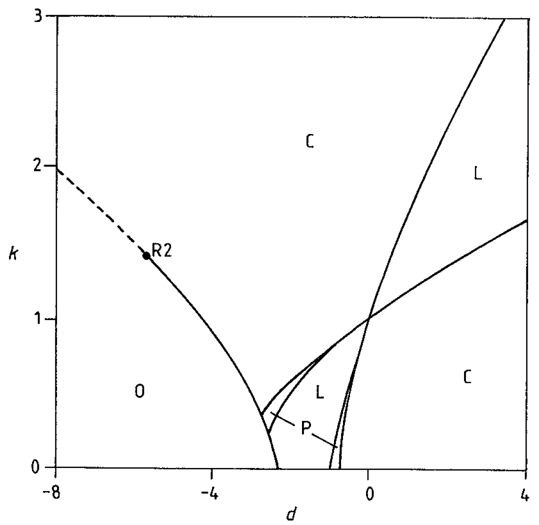

Fig. 3. Phase diagram at T=0 in the (k,d) plane. Full (broken) curves represent first (second) order phase transitions. R2 is a tri-critical point, O designates the disordered phase and P, C and L are ordered phases (ref. [38]).

The U_3X_4 systems are characterized by three four-fold symmetry axes on which the magnetic ions sit. This is explicitly reflected in the form of crystal–field Hamiltonian (15). From this fact and Eqs. (19) and (22), it follows that

$$\vec{n}_{2i} = \vec{n}_{2i-1} \;\text{ and }\; \mathcal{F}_{2i} = \mathcal{F}_{2i-1} \;\text{ for }\; i = 1, 2, 3.$$

Therefore from (20) we obtain that

$$\vec{m}_{2i} = \vec{m}_{2i-1},$$

i.e. our Hamiltonian describes a three-axial magnetic ordering.

The following analysis is similar to that of Khajehpour et al. [52] in the case of uniaxial ferromagnet with a crystal–field anisotropy.

To solve the system of self-consistent equations (19) and (20), one needs the eigenvalues of the site–Hamiltonian (23). In [38] this was done explicitly for the "spin"–value S=1. The results of the calculation are presented in fig. 3

At T=0 there are three possible solutions to Eqs. (19) and (20) that satisfy the necessary and sufficient conditions for a minimum of the free energy (18).

(1) The C-structure, i.e.

$$\vec{m}_1 = \vec{m}_2 = (u_c, v_c, v_c), \quad \vec{m}_3 = \vec{m}_4 = (v_c, u_c, v_c),$$
$$\vec{m}_5 = \vec{m}_6 = (v_c, v_c, u_c). \tag{24}$$

This is the same structure as described by (9) and presented in fig. 1.

(2) The L-structure, i.e.

$$\vec{m}_1 = \vec{m}_2 = \vec{m}_3 = \vec{m}_4 = (0, 0, v_L),$$
$$\vec{m}_5 = \vec{m}_6 = (0, 0, u_L)$$

which is the same as (10) and presented in fig. 2.

(3) The P-structure,

$$\vec{m}_1 = \vec{m}_2 = (u_P, v_P, 0), \quad \vec{m}_3 = \vec{m}_4 = (v_P, u_P, 0),$$
$$\vec{m}_5 = \vec{m}_6 = (w_P, w_P, 0). \tag{25}$$

Comparing the results of the MFA calculations at T=0 with those of the Landau theory we see that a new structure, i.e. the P-structure (25) has appeared. There is no contradiction, though, as the Landau theory is supposed to describe structures resulting in the continuous, i.e. second order transitions. The P-structure, as we shall see in Sec. 7 cannot be reached via continuous transition. From the phase diagram in fig. 3 we can see that there is only a very narrow region in which the P-structure would be stable. Such a structure has not yet been detected experimentally.

VI. BEHAVIOUR AT THE EXTERNAL MAGNETIC FIELD AT T=0 K

To calculate the behaviour of the magnetisation in an external magnetic field we followed the same procedure as in Sec. 5 while supplementing the Hamiltonian (16) with the Zeeman term

$$-\sum_{i,g_i,\alpha} H^\alpha S^\alpha_{g_i},$$

where $\vec{H}$ denotes the external magnetic field. The results of such numerical calculations are presented in fig. 4 for selected values of the interactions parameters.

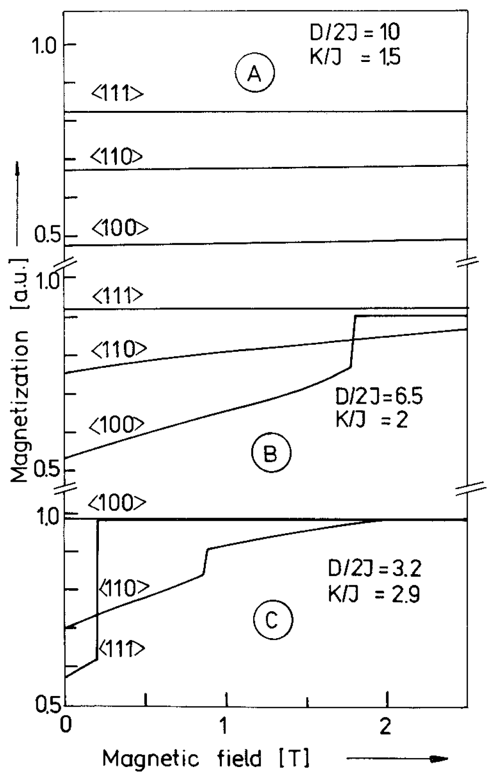

Fig. 4. Magnetisation along the principal axes predicted by spin model for three selected values of the crystal field and the exchange anisotropy (ref. [25]).

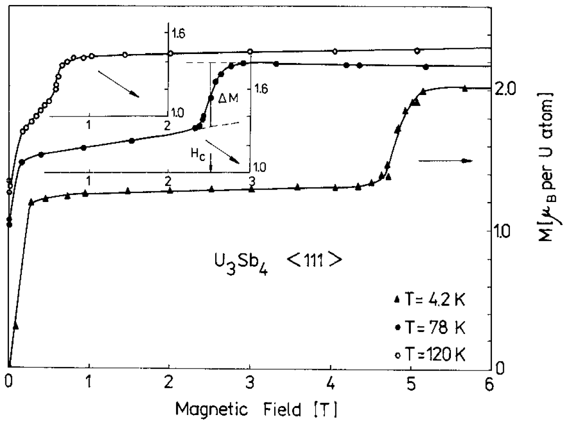

Fig. 5. Magnetisation curves for U_3Sb_4 along the $< 111 >$ axis at three different temperatures (ref. [26]).

The important feature of this diagram is that these theoretically calculated values of the magnetisation versus the external magnetic field closely resemble figures obtained experimentally. Thus in fig. 4A one can see the behaviour of the magnetisation as determined in [18, 21, 22] for U_3P_4 and in fig. 4B the dependence found in [19, 23, 24] for U_3As_4. Fig. 4C directly corresponds to the measurements on U_3Sb_4 reported in [25]. Especially the latter case, in view of the relative complexity of the magnetisation behaviour in external magnetic fields of various orientations, is worth noting. These results speak strongly in favour of the correctness of our model. The qualitative features of the results in fig. 4 can also be compared with the experimental curves presented in fig. 5 [26].

VII. NON-ZERO TEMPERATURE BEHAVIOUR

The complexity of the Hamiltonian (14), even in its mean–field simplified form (16) requires heavy numerical computation. Some analytic results can be obtained by exploiting high–temperature perturbation theory [38]. In this approach, the temperature–dependent molecular field term (17) can be treated as a small perturbation below the critical temperature. We may then calculate the free energies (22), and neglecting the higher order terms in the molecular fields (19), we obtain the following expression

$$\mathcal{F}_i = -f_0 - f_1 n_{i\perp}^2 - f_2 n_{i\|}^2 + \dots \tag{26}$$

where $\vec{n}_{i\perp}$ and $\vec{n}_{i\|}$ are the effective field components perpendicular and parallel, respectively to the direction of the crystal–field anisotropy axis in the ith sublattice, with

$$\vec{n}_{i\perp} + \vec{n}_{i\|} = \vec{n}_i, \quad f_0 = \frac{1}{b}\ln Z_0, \quad Z_0 = 1 + 2e^{\gamma},$$

$$\gamma = bd, \quad f_1 = e^{\gamma} - \frac{1}{dZ_0}, \quad f_2 = \frac{be^{\gamma}}{Z_0}.$$

Substituting Eq. (26) into Eq. (20) one obtains a system of homogeneous linear equations for the sublattice magnetisation $\vec{m}_i$. Equating the determinant to zero, one gets the equation for the Curie temperature

$$4f_1(8f_2 + k) - 1 = 0,$$

where $k = \frac{K}{J}$.

On the other hand, the question of the stability of various phases requires an examination of the higher order terms in the expansion. This was done in [38]. As the analysis confirms the results described in Sec. 3 and in Sec. 4 we shall illustrate them on phase diagrams given in fig. 6 and fig. 7. These are the phase diagrams in (τ, d) plane ($\tau = \frac{k_B T}{zJ}, \frac{D}{zJ}$) for selected values of the anisotropy constant $k = \frac{K}{J}$. Selection was after an examination of many such diagrams and the value $k = 2.9$ which is the same as in fig. 4C seems to be most relevant for U_3Sb_4.

For $k > 1$, the P phase does not occur at $T = 0$ and from all diagrams we have examined we could see that, when it appears, its stability range is very narrow (see, e.g., fig. 7) and it cannot be reached via a continuous (second order) transition. For k as high as in fig. 6 the P-phase disappears altogether.

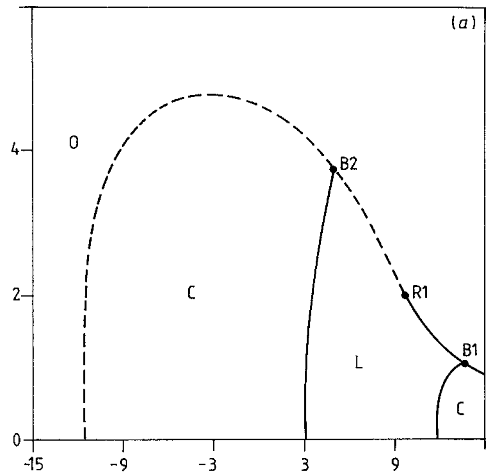

Fig. 6. Phase diagram in the (τ,d) plane for k=2.9. Full (broken) curves represent first- (second-) order phase transition lines (ref. [38]).

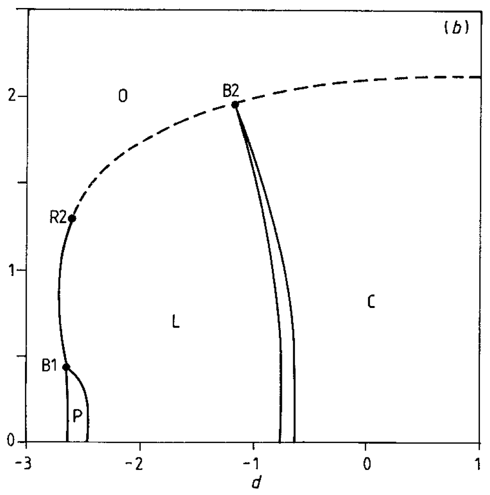

Fig. 7. Phase diagram in the (τ,d) plane for k=0.3. Full (broken) curves represent first- (second-) order phase transition lines (ref. [38]).

On the other hand the collinear phase L extends its range of stability as k increases. Both phases, L and C, can be reached from the paramagnetic phase either via first or second order phase transition. In other diagrams, which are not reproduced here (see [38]), for negative values of d a so-called re-entrant phase transition is possible (i.e. with decreasing τ we can first enter the ordered C-phase and then at still lower temperatures, returns to the disordered phase). Such a possibility was not found in the uniaxial case [52]. Another remarkable difference of this model and the uniaxial model is the decrease of the Curie temperature with increasing positive d, whereas in the latter T_c increases with positive d.

In fig. 5 are shown the experimental curves obtained while measuring the magnetisation of U_3Sb_4 versus external magnetic field. The data are obtained for the magnetic field directed along the $< 111 >$ direction at three different temperatures, i.e. 4.2 K, 78 K and 120 K. The measurements reveal the existence of a critical magnetic field, H_c, at which a spin reorientation transition takes place and a sudden jump, ΔM, of the magnetisation is observed. fig. 8 presents a comparison between the values of H_c and ΔM recorded at various temperatures and the values calculated for $k = 2.9$ and $d = 5.1$. These values were obtained by fitting the theoretical data to the experimental magnetisation along the $< 100 >$ axis at 4.2K. These results, in our opinion, confirm both the validity of the model and the existence of giant anisotropies arising from two different origins, namely the crystal field single ion anisotropy and the exchange effects between the nearest neighbours.

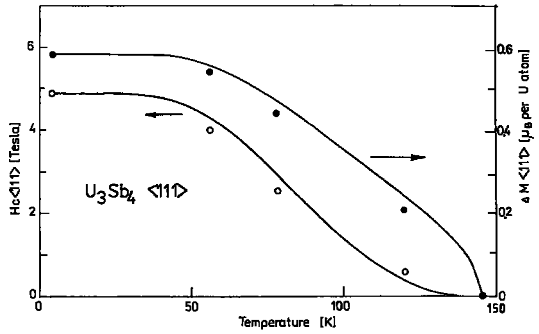

Fig. 8. Temperature dependence of the magnetisation jump ΔM, and the critical field H_c for the spin reorientation transition induced by the field along the $< 111 >$ axis. Points — experimental data; lines — data theoretically predicted and adjusted to the experimental points at 4.2 K (ref. [26]).

VIII. BEYOND THE MEAN FIELD: SPIN WAVE EXCITATIONS IN THE C AND L PHASE

On top of the fact that our model Hamiltonian (14) introduces three competing interactions of comparable magnitude (J, K and D) there is an additional difficulty, namely we have to deal with a six sublattice system as there are six magnetic ions in the unit cell. In such a case it is difficult to discuss the elementary excitations even in the harmonic approximation [53]. However, as it has been shown by Kaganov and Chubukov [54], the single–ion crystal field interaction leads to strong spin–fluctuations, which may profoundly affect both the magnetic ordering and excitations. The molecular field equations for the spin orientations turn out not to be sufficient and one has to go beyond the harmonic approximation to discuss the stability of the system. Also from this point of view our model provides an interesting example to check the validity of the Kaganov–Chubukov theory. We work in the spin–wave approximation, i.e. we look for the spin–waves energies upon the diagonalisation of the bilinear spin–wave Hamiltonian. Because we are dealing with six sublattices we transform the spin operators in (14) to a local coordinate system

$$S^{\alpha}_{g(p)} = \rho^{\alpha}_{p}\sigma^{z}_{g(p)} + U^{\alpha}_{p}\sigma^{+}_{g(p)} + \bar{U}^{\alpha}_{p}\sigma^{-}_{g(p)},$$

where ρ^{α}_{p} denotes the unit vector along the easy axis in the pth sublattice and dash means complex conjugate

$$U^{x}_{p} = -\frac{1}{4}e^{i\Phi_p}(1+\rho^{z}_{p}) + \frac{1}{4}e^{-i\Phi_p}(1-\rho^{z}_{p}),$$

$$U^{y}_{p} = \frac{i}{4}\left[e^{i\Phi_p}(1+\rho^{z}_{p}) + e^{-i\Phi_p}(1-\rho^{z}_{p})\right],$$

$$U^{z}_{p} = \frac{1}{2}\sqrt{1-(\rho^{z}_{p})^2}, \quad \tan\Phi_p = \frac{\rho^{y}_{p}}{\rho^{x}_{p}}.$$

A standard Holstein–Primakoff transformation transforms the Hamiltonian (14) into the form

$$\tilde{\mathcal{H}} = \tilde{\mathcal{H}}_0 + \tilde{\mathcal{H}}_1 + \tilde{\mathcal{H}}_2 + \dots\ ,$$

$$\tilde{\mathcal{H}}_0 = -S\sum_{p<q}\sum_{\alpha}\frac{J^{\alpha\alpha}_{pq}}{J}\rho^{\alpha}_{p}\rho^{\alpha}_{q} - d\sum_{p}\left\{S\left(\rho^{\mu(p)}_{p}\right)^2 + 2\left|U^{\mu(p)}_{p}\right|^2\right\} - S\sum_{p}\sum_{\alpha}h^{\alpha}\rho^{\alpha}_{p},$$

$$\begin{aligned}\tilde{\mathcal{H}}_1 \ = \ & -\sqrt{\frac{2S}{N}}\Big\{\sum_{p<q}\sum_{\alpha}\frac{J^{\alpha\alpha}_{pq}}{J}\left(U^{\alpha}_{p}\rho^{\alpha}_{q}a^{(p)}_{0} + U^{\alpha}_{q}\rho^{\alpha}_{p}a^{(q)}_{0}\right) + 2\tilde{d}\sum_{p}\rho^{\mu(p)}_{p}U^{\mu(p)}_{p}a^{(p)}_{0}\\ & + \sum_{p}\sum_{\alpha}h^{\alpha}U^{\alpha}_{p}a^{(p)}_{0} + H.c.\Big\},\end{aligned}$$

$$\begin{aligned}
\tilde{\mathcal{H}}_2 \;=\; & -\frac{1}{N}\sum_{\mathbf{k}}\Big[\sum_{p<q}\sum_{\alpha}\frac{J^{\alpha\alpha}_{pq}}{J}\Big\{-\rho^{\alpha}_{p}\rho^{\alpha}_{q}a^{(p)+}_{\mathbf{k}}a^{(p)}_{\mathbf{k}}+2\bar{U}^{\alpha}_{p}U^{\alpha}_{q}\gamma^{pq}_{-\mathbf{k}}a^{(p)+}_{\mathbf{k}}a^{(q)}_{\mathbf{k}} \\
& +U^{\alpha}_{p}U^{\alpha}_{q}\left(\gamma^{pq}_{\mathbf{k}}a^{(p)}_{\mathbf{k}}a^{(q)}_{-\mathbf{k}}+\gamma^{pq}_{-\mathbf{k}}a^{(p)}_{-\mathbf{k}}a^{(q)}_{\mathbf{k}}\right)+H.c.\Big\} \\
& +\sum_{p}\Big\{2\tilde{d}\Big(2|U^{\mu(p)}_{p}|^2-\left(\rho^{\mu(p)}_{p}\right)^2\Big)a^{(p)+}_{\mathbf{k}}a^{(p)}_{\mathbf{k}} \\
& +2d\sqrt{1-1/2S}\Big((U^{\mu(p)}_{p})^2a^{(p)}_{\mathbf{k}}a^{(p)}_{-\mathbf{k}}+H.c.\Big)\Big\}+\sum_{p}\sum_{\alpha}h^{\alpha}\rho^{\alpha}_{p}a^{(p)+}_{\mathbf{k}}a^{(p)}_{\mathbf{k}}\Big],
\end{aligned}$$

where

$\tilde{\mathcal{H}}=\frac{\mathcal{H}}{zSNJ}$, $h^{\alpha}=\frac{H^{\alpha}}{zSJ}$, $\kappa=\frac{K}{J}$, $d=\frac{D}{zJ}$, $\gamma^{pq}_{\mathbf{k}}=\frac{1}{2}\sum_{g(p),g(q)}\exp\{i\vec{k}(\vec{r}_{g(q)}-\vec{r}_{g(p)})\}$, $\tilde{d}=d(1-1/2S)$.

The ground state is determined by eliminating the linear term in the bosonic operators, i.e.

$$\tilde{\mathcal{H}}_1=0 \tag{27}$$

then the spin wave energies should be obtained by diagonalisation of the bilinear operator $\tilde{\mathcal{H}}_2$. It can be checked that both the C and the L structures fulfill the condition (27). For the C-structure (24), putting

$$\vec{\rho}_1=\vec{\rho}_2=\begin{pmatrix}u\\v\\v\end{pmatrix},\quad \vec{\rho}_3=\vec{\rho}_4=\begin{pmatrix}v\\u\\v\end{pmatrix},\quad \vec{\rho}_5=\vec{\rho}_6=\begin{pmatrix}v\\v\\u\end{pmatrix},$$

and considering the external field $\vec{h}$ along the (1,1,1) direction, $\vec{h}=(h,h,h)$, the condition (27) gives

$$\left.\begin{aligned}4v^2-2u^2+\delta uv+h(v-u)&=0\\ u^2+2v^2&=1\end{aligned}\right\},$$

where $\delta=\tilde{d}-\kappa$. For $h=0$ we have two types of solution

$$v_1=\pm\frac{1}{2}\Delta_{(-)}\;,u_1=\pm\frac{1}{2}\sqrt{2}\Delta_{(+)}\;, \tag{28}$$

$$v_2=\pm\frac{1}{2}\Delta_{(+)}\;,u_2=\mp\frac{1}{2}\sqrt{2}\Delta_{(-)}\;, \tag{29}$$

where

$$\Delta_{(\pm)}=\sqrt{1\pm\frac{\delta}{\sqrt{8+\delta^2}}}\,.$$

The solution (29) does not correspond to the isotropic case and therefore we are left with (28) to consider. It gives the following angle of non-collinearity

$$\tan 2\theta=2\sqrt{2}\frac{1+\delta}{8-\delta}.$$

Typical numerical results for the case of the non-collinear ordering of U_3P_4 are presented in fig. 9 for S=2, κ=2, d=3.5 and h=0. We observe a gap at the Γ point and the 6-fold degeneracy of the spectrum at H-point of the Brillouin zone. There is also a $\vec{k}$-independent branch along the Δ-line. Futher calculation reveal that the gap is not a monotonic function of d and that the spectrum can become gapless for some values of d. This happens at the borders of stability of the phase.

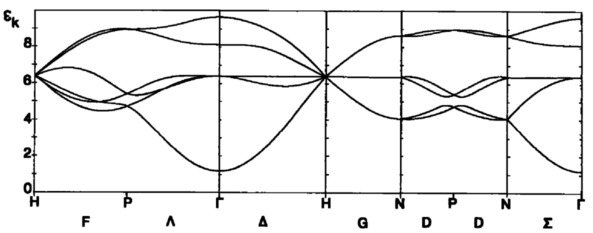

Fig. 9. Energy spectrum of non-collinear ferromagnet like U_3P_4 for S=2, κ=2, d=3.5 and h=0. The symmetry points of the Brillouin zone of the body centred cubic lattice: Γ=(0,0,0), H=$(2\pi/a)$(0,1,0), P=(π/a)(1,1,1), N=(π/a)(1,1,0) (ref. [39]).

We found that the lowest energy branch, near the Γ-point of the BZ is parabolic. This is important in view of a suggestion made by Markowski et al. [55] that a $T^{3/4}$-power law behaviour of the magnetisation at low temperatures should be expected. However other experiments (see, e.g., ref. [56]) did not observe any such anomalous behaviour.

The problem of spin waves in the L-structure is more complex. Our calculations in the harmonic approximation lead to small negative spin wave energies. This instability is indeed very small and e.g. for S=2, κ=2, d=1.333 the lowest energy value is $\epsilon_\Gamma = -0.002$. This instability can be easily removed by a small external field along the easy axis. The spectra obtained at such a field are noticeably different from these for U_3P_4. For example: the H-point degeneracy is removed and some new $\vec{k}$-independent branches appear.

However, one needs to look for a better remedy than the external field. Two ways out of these difficulties will be considered. One is to check the Kaganov–Chubukov conjecture [54] that it is due to large spin–fluctuations caused by the single–ion term and calculate the higher order corrections. The other is to consider some additional relativistic anisotropy, like, e.g., Eq. (13). The latter, however, will probably lead first to a small non-collinearity of the ground state as has been suggested by Sandratskii and Kübler [57] and in agreement with Eq. (11).

IX. CONCLUDING REMARKS

We have presented here a Heisenberg–type spin model to describe the magnetic properties of the U_3X_4 compounds. This model has been derived using the Landau symmetry analysis of the ordered structures and examining the invariant contributions to the Landau thermodynamic potential. In the U_3X_4 case we have to deal with a complex form where couplings between the different fields belonging to the same irreducible representation of the Hamiltonian symmetry group should be considered. From these bilinear couplings three important contributions are selected: (1) the usual exchange nearest–neighbour Hamiltonian, (2) single–ion crystal field anisotropy and (3) anisotropic exchange between nearest–neighbours. These three types of interactions turn out to be sufficient to describe various effects observed in U_3X_4 compounds. Upon examining the magnetisation behaviour under the external magnetic field we can conclude that these interactions are of comparable magnitude, i.e. we are dealing with giant anisotropies. The question of the magnetic structure is solved consistently and the results are in excellent agreement with the modern neutron diffraction investigations. The model predicted the non-collinear structure of U_3P_4 and U_3As_4 and also the collinear ferrimagnetism of U_3Sb_4 and U_3Bi_4.

A more fundamental approach of Sandratskii and Kübler, based directly on the electronic structure of the U_3X_4 compounds suggests that perhaps more subtle relativistic interactions may be important and lead to a small perpendicular antiferromagnetic arrangement. If this is the case then one should reconsider other relativistic contributions to the Landau potential that were neglected in constructing our model Hamiltonian. Then the influence of such terms would be discussed. It is however surprising that in the Sandratskii and Kübler approach the giant exchange anisotropy K, which is important to understand the behaviour of the U_3X_4 compounds in terms of our Hamiltonian, is missing.

ACKNOWLEDGMENTS

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МАҐНЕТИЗМ В УРАНОВИХ СПОЛУКАХ КРИСТАЛІЧНОЇ СТРУКТУРИ Th_3P_4

Ч. Олекси, Є. Пшистава
Інститут теоретичної фізики Вроцлавського університету,
Польща, 50–204, Вроцлав, пл. Макса Борна, 9

Обговорюється питання магнетизму сполук U_3X_4 (X=P, As, Sb, Bi) в термінах запропонованої авторами у 1987 році моделі типу Гайзенберга. Остання включає конкуруючі однойонні та анізотропні обмінні взаємодії порівняльної величини. Модель виведена на основі аналізу симетрії структур та теорії фазових переходів Ландау. Представлено результати обчислень у наближенні середнього поля для опису магнетних структур та їх термодинамічної поведінки.