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ON SPIN WAVES IN U_3Sb_4

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The spin wave excitations in the magnetic structure of U_3Sb_4 are analysed using the Onufrieva method. This analysis removes the difficulty noted in earlier works with negative magnon energies. The calculations were performed for spin $S = 1$ along some lines of the Brillouin zone.

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 been attracting experimental attention for 35 years [1–11]. These materials are highly anisotropic and their ordered phases are complicated multi–axial structures. U_3P_4 and U_3As_4 were found [8] to be non–collinear three–axial ferromagnets in agreement with the earlier theoretical prediction [12,13]. Recent neutron diffraction experiment on U_3Sb_4 and U_3Bi_4 [11] defined their magnetic structures to be collinear with two magnetic sublattices.

The multi–axial structures with single–ion crystal field anisotropy and exchange interaction presented a serious theoretical problem. The first effective spin model proposed by Przystawa [14,15] described only non–collinear ordered structure. Symmetry analysis of possible magnetic interactions (see e.g., [16]) have revealed the existence of an unusual type of the exchange anisotropy in these perfect cubic structures. The Heisenberg–type model with competitive exchange anisotropy and crystal field anisotropy interactions has been used to study ground–state phase diagrams [9], phase transitions [10,17], and spin–wave excitations [18]. A good agreement with experimental data can be achieved if competitive interactions are strong enough and of comparable magnitude [10]. The description of spin waves in a collinear phase of U_3Sb_4 is a very difficult problem. Some energy excitations calculated in the harmonic approximation become negative [18]. This is due to a large spin fluctuations caused by the crystal field interaction [19], thus higher order anharmonic terms in bosonic operators should be accounted for. However in case of U_3Sb_4 the crystal field anisotropy cannot be treated as small with respect to exchange interaction [10]. In this paper we use an alternative method, proposed by Onufrieva [20–22], to study the energy excitations in U_3Sb_4. In this method the spin operators are replaced by the SU(2S+1) Lie algebra generators in order to diagonalize the single–ion part of the Hamiltonian via unitary transformation. Next the quasi–particle idea is introduced by using generalized Holstein–Primakoff (or Dyson) transformation.

II. CRYSTAL AND MAGNETIC STRUCTURE OF U_3X_4

All the U_3X_4 compounds crystallize in the body centred cubic structure of Th_3P_4 with the cubic space group $I\bar{4}3d$ – Td^6. Magnetic structures of these compounds can be described by six spin vectors $\mathbf{S}_1, \ldots, \mathbf{S}_6$ situated at uranium ions positions in the crystallographic unit cell:

$$\begin{aligned} &\mathbf{r}_1 = (\tfrac{3}{8}, 0, \tfrac{1}{4}),\ \mathbf{r}_2 = (\tfrac{1}{8}, 0, \tfrac{3}{4}),\ \mathbf{r}_3 = (\tfrac{1}{4}, \tfrac{3}{8}, 0),\\ &\mathbf{r}_4 = (\tfrac{3}{4}, \tfrac{1}{8}, 0),\ \mathbf{r}_5 = (0, \tfrac{1}{4}, \tfrac{3}{8}),\ \mathbf{r}_6 = (0, \tfrac{3}{4}, \tfrac{1}{8}). \end{aligned} \tag{1}$$

Landau analysis of the phase transitions in U_3X_4 [23,24] predicted two ferromagnetic spin structures. The first is a noncollinear six–axial structure

$$\begin{aligned} &\mathbf{S}_1 = \begin{pmatrix} u \\ v \\ w \end{pmatrix},\ \mathbf{S}_2 = \begin{pmatrix} u \\ w \\ v \end{pmatrix},\ \mathbf{S}_3 = \begin{pmatrix} w \\ u \\ v \end{pmatrix},\\ &\mathbf{S}_4 = \begin{pmatrix} v \\ u \\ w \end{pmatrix},\ \mathbf{S}_5 = \begin{pmatrix} v \\ w \\ u \end{pmatrix},\ \mathbf{S}_6 = \begin{pmatrix} w \\ v \\ u \end{pmatrix} \end{aligned} \tag{2}$$

with the resulting magnetic moment along the [111] direction. This type of structure (with $w = v$) was determined in [8] for U_3P_4 and U_3As_4.

The second possible structure is :

$$\begin{aligned} &\mathbf{S}_1 = \begin{pmatrix} 0 \\ -\gamma \\ \rho \end{pmatrix},\quad \mathbf{S}_2 = \begin{pmatrix} 0 \\ \gamma \\ \rho \end{pmatrix},\quad \mathbf{S}_3 = \begin{pmatrix} \gamma \\ 0 \\ \rho \end{pmatrix},\\ &\mathbf{S}_4 = \begin{pmatrix} -\gamma \\ 0 \\ \rho \end{pmatrix},\ \mathbf{S}_5 = \mathbf{S}_6 = \begin{pmatrix} 0 \\ 0 \\ \sigma \end{pmatrix}, \end{aligned} \tag{3}$$

i.e., the structure with the resulting magnetic moment along the [001] direction. This structure with $\gamma = 0$ was determined recently in neutron diffraction experiment [11] for U_3Sb_4 and U_3Bi_4. The same magnetic structure as eq. (2) and eq. (3) has been recently obtained in the first–principle calculation [25–27].

The difference between theoretical prediction and experimentally determined magnetic structure has been explained [16,24] by discussing the contributions to the Landau potential coming from "exchange forces" and "relativistic forces" [28]. The latter are assumed to be weak. Neglecting relativistic terms in the Landau potential leads to structure (2) with $w = v$ and structure (3) with $\gamma = 0$. This indicates that relativistic corrections to magnetic structures are very small. It was confirmed experimentally [11] for U_3Sb_4 where finding the deflection of magnetic moments from the [001] direction was beyond the experimental possibilities. In further analysis we shall also neglect the relativistic forces mainly in order to simplify this complicated calculation.

III. THE MODEL HAMILTONIAN

A model Hamiltonian to describe the magnetically ordered state of uranium compounds was constructed by making use of the symmetry analysis and Landau theory of the phase transition (for more details see Ref. [16]).

It contains spin–spin interactions of the following type: (1) the nearest–neighbour exchange interactions, (2) the crystal–field single–ion term, and (3) the nearest–neighbour "exchange–type" anisotropy.

$$\mathcal{H} = \mathcal{H}_I + \mathcal{H}_{CF}, \tag{4}$$

$$\mathcal{H}_{\mathcal{I}} = -\frac{1}{2}\sum_{i,j}\sum_{g_i,g_j}\sum_{\alpha} J^{\alpha\alpha}_{g_i g_j} S^{\alpha}_{g_i} S^{\alpha}_{g_j} \tag{5}$$

where

$S^{\alpha}_{g_i}$ is the αth component of the spin operator at the g–th site in the sublattice (i=1,2, ...,6)

$J^{\alpha\alpha}_{g_i g_j}$ is the anisotropic interaction tensor,

$\mathcal{H}_{CF}$ is the crystal field Hamiltonian, viz.

$$\mathcal{H}_{CF} = -D\sum_{i,g_i} \hat{Q}_{g_i} \tag{6}$$

where

$$\hat{Q}_{g_i} = \begin{cases} (S^x_{g_i})^2 & \text{for } i = 1,2 \\ (S^y_{g_i})^2 & \text{for } i = 3,4 \\ (S^z_{g_i})^2 & \text{for } i = 5,6 \end{cases} .$$

The exchange interaction tensor can be specified as

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

$$J_{12} = J_{34} = J_{56} = 0,$$

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

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

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

It is worth noting that our Hamiltonian contains 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.

IV. MEAN–FIELD APPROXIMATION

We start the analysis of magnetic structure described by the model Hamiltonian, eq. (4), using the Molecular Field Approximation (MFA). In the MFA Hamiltonian the crystal field term is treated exactly and exchange type terms are linearized by introducing the effective field $\mathbf{n}_i$ acting on the magnetic ions on the sublattice i

$$\tilde{\mathcal{H}} = \mathcal{H}_{CF} - \sum_{i,g_i,\alpha} n^{\alpha}_i S^{\alpha}_{g_i}. \tag{7}$$

Numerical calculation of the sublattice magnetisation $\mathbf{m}_i = \langle S_i \rangle$ performed for the spin $S = 1$ and the positive crystal field constant D at the temperature $T = 0$ leads to two solutions.
(1) The C–structure

$$\begin{aligned} \mathbf{m}_1 = \mathbf{m}_2 &= (u_c, v_c, v_c), \\ \mathbf{m}_3 = \mathbf{m}_4 &= (v_c, u_c, v_c), \\ \mathbf{m}_5 = \mathbf{m}_6 &= (v_c, v_c, u_c). \end{aligned} \tag{8}$$

This is the structure described by eq. (2) with $w = v$.
(2) The L–structure

$$\mathbf{m}_1 = \mathbf{m}_2 = \mathbf{m}_3 = \mathbf{m}_4 = (0, 0, v_L), \tag{9}$$

$$\mathbf{m}_5 = \mathbf{m}_6 = (0, 0, u_L),$$

which corresponds to structure (3) with $\gamma = 0$. It would be possible to obtain exactly the same magnetic structures as those in Section II by the inclusion into the Hamiltonian eq. (4) of a"broken" Dzialoshinsky–Moriya term [16] allowed by the symmetry.

The phase diagram at $T = 0$ presented in fig. 1 shows that the L–structure is stable when the amplitudes of anisotropic exchange interaction $\kappa = K/J$ and the crystal field anisotropy $d = D/2J$ are of comparable magnitude. It is worth noting that for negative values of d the third solution with a planar magnetic structure is possible [17].

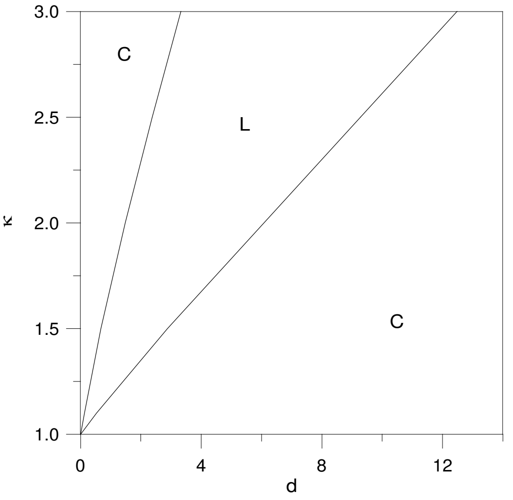

Fig. 1. The phase diagram at $T = 0$. L and C denote ordered phases.

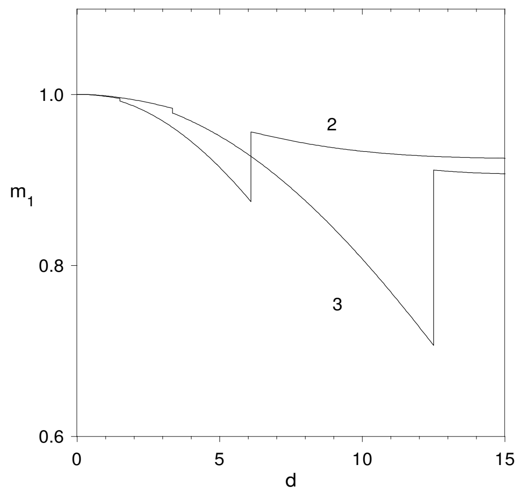

Fig. 2. Reduction of magnetisation on sublattice 1 as a function of d for two values $\kappa = 2, 3$. The lower part of each curve corresponds to the magnetisation in the L–structure.

The presence of the crystal field anisotropy determines that sublattice magnetisations do not saturate, i.e. $m_i < 1$, in the C–structure and in the L–structure on sublattices $1, \ldots, 4$. The reduction of magnetisation increases with the crystal field constant (see fig. 2) and it is high for the L–structure, e.g., at the point $\kappa = 3$ and $d = 12.5$ the magnetisation $m_1 = 0.7$ is reduced by 30%. We think this is the main reason that the harmonic spin wave approximation does not work in U_3Sb_4.

V. SPIN WAVE APPROACH TO U_3SB_4

It is known that in the presence of single–ion anisotropy in the spin Hamiltonian the standard Holstein–Primakoff (H–P) or Dyson transformations may lead to unphysical results [19]. One way to remove this trouble is taking into account higher order anharmonic terms in the bosonic Hamiltonian. However, such an approach is limited to small single–ion anisotropy constants in comparison with the exchange constants. The other way, proposed by Onufrieva [20–22], replaces the spin operators in a Hamiltonian by the $SU(2S + 1)$ Lie algebra generators in order to diagonalize the single–ion part of the Hamiltonian via a unitary transformation. Then, the quasi–particles idea is used by properly introducing a generalized Holstein–Primakoff (Dyson) transformation. The latter describes excited states by $2S$ independent bosonic operators. In this approach there is no limitation in the magnitude of the anisotropy constant.

In earlier work [18] we demonstrated that magnon excitations in non–collinear magnets U_3P_4 and U_3As_4 can be studied by making use of a standard H–P transformation, particularly for the spin $S \geq 2$. On the other hand, this method leads to unphysical results in case of the collinear magnet U_3Sb_4 for any value of the spin S. It has been recently shown [29] that introducing the Dzialoshinski–Moriya like term to Hamiltonian leads to small noncollinearity in the ground state solutions in agreement with results of symmetry analysis [24] as suggested by Sandratskii and Kübler [27]. However, in this case the magnon excitations described by the standard H–P transformation are also unphysical. Our results obtained in the MFA show that in the case of U_3Sb_4 the value of the single–ion anisotropy D is larger than the exchange interaction constants J and K (see fig. 1). Therefore we shall use here the method proposed by Onufrieva [20–22] for the spin $S = 1$, and our analysis of energy excitations will be performed for U_3Sb_4.

There are 8 independent operator of the SU(3) algebra, $S^z, S^{\pm}, O^0, O^{\pm 1}, O^{\pm 2}$,

$$S^{\pm} = \mp\frac{1}{\sqrt{2}}(S^x \pm iS^y), O^0 = (S^z)^2 - \frac{2}{3}I, O^{\pm 1} = -(S^z S^{\pm} + S^{\pm} S^z), O^{\pm 2} = (S^{\pm})^2.$$

The matrix form of these operators can be written as

$$S^z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad S^+ = \begin{pmatrix} 0 & -1 & 0 \\ 0 & 0 & -1 \\ 0 & 0 & 0 \end{pmatrix}, \quad S^- = \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \tag{10}$$

$$O^0 = \frac{1}{3}\begin{pmatrix} 1 & 0 & 0 \\ 0 & -2 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad O^1 = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & -1 \\ 0 & 0 & 0 \end{pmatrix}, \quad O^{-1} = \begin{pmatrix} 0 & 0 & 0 \\ -1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \tag{11}$$

$$O^2 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad O^{-2} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}. \tag{12}$$

We divide the Hamiltonian (4) into single–ion and interaction parts by introducing

$$\delta S^\alpha = S^\alpha - \langle S^\alpha \rangle. \tag{13}$$

Then using the $SU(3)$ generators the Hamiltonian can be written as

$$\mathcal{H} = E_0 + \mathcal{H}_0 + \mathcal{H}_{int}, \tag{14}$$

where

$$\mathcal{H}_0 = -\sum_i \mathcal{H}_{0i}, \quad \mathcal{H}_{0i} = -\sum_{f_i} \Big(h_i S^z_{f_i} + c_i O^0_{f_i} + g_i (O^{-2}_{f_i} + O^2_{f_i}) \Big), \tag{15}$$

$$\mathcal{H}_{int} = -\frac{1}{2} \sum_{i,j} \sum_{f_i, f_j} \sum_{\alpha,\beta} \tilde{J}^{\alpha\beta}_{f_i f_j} \delta S^\alpha_{f_i} \delta S^\beta_{f_j}. \tag{16}$$

According to the MFA solution (9) corresponding to the U_3Sb_4 case we can assume $\langle S^\pm \rangle = 0$ obtaining

$$h_1 = h_2 = 2K(\langle S^z_3 \rangle + \langle S^z_4 \rangle) + 2J(\langle S^z_5 \rangle + \langle S^z_6 \rangle),$$

$$h_3 = h_4 = 2K(\langle S^z_1 \rangle + \langle S^z_2 \rangle) + 2J(\langle S^z_5 \rangle + \langle S^z_6 \rangle),$$

$$h_5 = h_6 = 2J(\langle S^z_1 \rangle + \langle S^z_2 \rangle + \langle S^z_3 \rangle + \langle S^z_4 \rangle), \tag{17}$$

$$c_i = -\frac{1}{2} D \quad \text{for } \ i = 1, \ldots, 4, \quad c_5 = c_6 = D,$$

$$g_1 = g_2 = \frac{1}{2} D, \quad g_3 = g_4 = -\frac{1}{2} D, \quad g_5 = g_6 = 0,$$

$$\tilde{J}^{++}_{15} = \tilde{J}^{--}_{15} = \frac{1}{2}(J - K), \quad \tilde{J}^{++}_{35} = \tilde{J}^{--}_{35} = -\frac{1}{2}(J - K), \quad \tilde{J}^{+-}_{15} = \tilde{J}^{+-}_{35} = -\frac{1}{2}(J + K),$$

$$\tilde{J}^{-+}_{13} = -J, \quad \tilde{J}^{--}_{13} = \tilde{J}^{++}_{13} = 0, \quad \tilde{J}^{zz}_{15} = \tilde{J}^{zz}_{35} = J, \quad \tilde{J}^{zz}_{13} = K.$$

The remaining elements can be obtained from the symmetry of the tensor $\tilde{J}$:

$$\tilde{J}^{\alpha\beta}_{2iq} = \tilde{J}^{\alpha\beta}_{2i-1q}, \quad \tilde{J}^{\alpha\beta}_{pq} = \quad \tilde{J}^{\beta\alpha}_{pq} = \tilde{J}^{\alpha\beta}_{qp},$$

$$\tilde{J}^{\alpha\beta}_{pp} = 0 \quad \tilde{J}^{z\pm}_{pq} = 0.$$

The single–ion Hamiltonian can be easily diagonalized via transformations Ψ_p, where $p = 1, 2, 3, 4$ denotes the sublattice number:

$$\mathcal{H}_{0p}(\phi_p) = \Psi_p \mathcal{H}_{0p} \Psi_p^{-1}, \tag{18}$$

where

$$\Psi_p = \begin{pmatrix} \cos\phi_p & 0 & \sin\phi_p \\ 0 & 1 & 0 \\ -\sin\phi_p & 0 & \cos\phi_p \end{pmatrix}. \tag{19}$$

These transformations change the operator A into $A(\phi_p) = \Psi_p A \Psi_p^{-1}$ which can be expressed in terms of the $SU(3)$ generators. Thus, the operators occurring in Hamiltonian (18) have the following representation:

$$O^0(\phi_p) = O^0, \tag{20}$$

$$S^\pm(\phi_p) = S^\pm \cos\phi_p + O^{\mp 1} \sin\phi_p, \tag{21}$$

$$\begin{pmatrix} S^z(\phi_p) \\ O^2(\phi_p) \\ O^{-2}(\phi_p) \end{pmatrix} = \mathcal{R}_p \begin{pmatrix} S^z \\ O^2 \\ O^{-2} \end{pmatrix}, \tag{22}$$

where

$$\mathcal{R}_p = \begin{pmatrix} \cos 2\phi_p & -\sin 2\phi_p & -\sin 2\phi_p \\ \frac{1}{2}\sin 2\phi_p & \cos^2\phi_p & -\sin^2\phi_p \\ \frac{1}{2}\sin 2\phi_p & -\sin^2\phi_p & \cos^2\phi_p \end{pmatrix}. \tag{23}$$

Now the sublattice Hamiltonian can be written as

$$\mathcal{H}_{0p}(\phi_p) = -\sum_{f_p} \left(\tilde{h}_p S^z_{f_p} + c_p O^0_{f_p} + \tilde{g}_p (O^{-2}_{f_p} + O^2_{f_p}) \right) \tag{24}$$

where

$$\begin{matrix} \tilde{h}_p = h_p \cos 2\phi_p + g_p \sin 2\phi_p \\ \tilde{g}_p = h_p \sin 2\phi_p - g_p \cos 2\phi_p \end{matrix} \quad \text{for } p = 1, \ldots, 4, \tag{25}$$

$$\tilde{h}_p = h_p, \quad \tilde{g}_p = 0 \qquad \text{for } p = 5, 6. \tag{26}$$

The off–diagonal terms in Hamiltonian (24) vanish when

$$\tilde{g}_p = 0 \qquad \text{for } p = 1, \ldots, 6. \tag{27}$$

Mean values of the operators occuring in eqs. (17) will be calculated in the single–ion approximation:

$$\langle A_p \rangle^0 = \frac{Tr\{A_p \exp(-\mathcal{H}_{0p}/k_B T)\}}{Tr\{\exp(-\mathcal{H}_{0p}/k_B T)\}}. \tag{28}$$

In the transformed system the mean value $\langle A_p \rangle^0_{\phi_p}$ is calculated using eq. (28) with $\mathcal{H}_{0p}(\phi_p)$. Since $\mathcal{H}_{0p}(\phi_p)$ is diagonal then there are only two non–zero mean values:

$$\sigma_p = \langle S^z_p \rangle^0_{\phi_p},$$

$$\lambda_p = \langle 3 O^0_p \rangle^0_{\phi_p}.$$

Using the invariance of the trace with respect to unitary transformations the mean values of the operators $\langle A_p \rangle^0$ in the initial system can be expressed via σ_p and λ_p. It is worth noting that at temperature $T = 0$ there exist the solution $\sigma_p = 1$, $\lambda_p = 1$ for $p = 1, \ldots, 6$, which indicate that magnetisation reaches the saturation value in the transformed system. On the other hand the sublattice magnetisations in the initial system

$$\langle S^z_p \rangle^0 = \begin{cases} \sigma_p \cos 2\alpha & \text{for } p = 1, \ldots, 4 \\ \sigma_p & \text{for } p = 5, 6 \end{cases} \tag{29}$$

do not saturate in the four sublattices.

The solution of the system of equations (27) is the following

$$\phi_1 = \phi_2 = -\phi_3 = -\phi_4 = \alpha, \tag{30}$$

where α is the root of the non–linear equation

$$(K \cos 2\alpha + J) \sin 2\alpha - \frac{D}{8} \cos 2\alpha = 0. \tag{31}$$

Sublattice transformations (19) change the form of the $\mathcal{H}_{int}(\alpha)$ which now depends on all generators with the exception of O^0.

The transition from the spin operators to boson operators is performed via non–standard Holstein–Primakoff (or Dyson) transformation [21]. First we express the $SU(3)$ generators in the terms of the Hubbard operators X^{pq} which describe a transition from the single–ion eigenstate $|q\rangle$ of the S^z operator to the eigenstate $|p\rangle$. The matrix representation of X^{pq} has only one element (the pq–element) different from zero. Here the $|1\rangle$ is assumed to be the ground state:

$$S^- = X^{01} + X^{-10}, \quad S^+ = -X^{10} - X^{0-1}, \tag{32}$$

$$S^Z = 1 - X^{00} - 2X^{-1-1}, \quad O^0 = \frac{1}{3} - X^{00}, \tag{33}$$

$$O^{-1} = -X^{01} + X^{-10}, \quad O^{+1} = X^{10} - X^{0-1}, \tag{34}$$

$$O^{-2} = X^{-11}, \quad O^{+2} = X^{1-1}. \tag{35}$$

The Hubbard operators are expressed by means of two sets of boson operators (a^+, a) and (b^+, b) [21]:

$$X^{01} = a^+ A, \quad X^{-11} = b^+ A, \quad X^{00} = a^+ a, \tag{36}$$
$$X^{-1-1} = b^+ b, \quad X^{0-1} = a^+ b,$$

where

$$A = \sqrt{1 - a^+ a - b^+ b}. \tag{37}$$

The a, a^+ and b, b^+ operators can be treated as boson operators only approximately when the number of excited magnons is very small. This limits our discussion of the spin wave excitations to a low temperature region. In constructing the magnon effective Hamiltonian we take into account only the square terms of operators a^+, a, b^+, b. The contribution from $\mathcal{H}_{int}(\alpha)$ can be easily obtained by using linearized form of the spin operators:

$$S^- = a^+, \quad S^+ = -a, \quad O^{-1} = -a^+, \quad O^1 = a, \tag{38}$$
$$O^{-2} = b^+, \quad O^2 = b, \quad \delta S^z = 0.$$

Finally, we can write the harmonic part of the boson Hamiltonian as

$$\mathcal{H}_{SWA} = \sum_{j,f_j} \left(\left(\tilde{h}_j + c_j \right) a^+{}_{f_j} a_{f_j} + 2\tilde{h}_j b^+{}_{f_j} b_{f_j} \right) + \tag{39}$$
$$- \frac{1}{2} \sum_{i,j} \sum_{f_i,f_j} \left(V_{ij}^{+-} a^+{}_{f_i} a_{f_j} + V_{ij}^{--} a_{f_i} a_{f_j} + U_{ij}^{+-} b^+{}_{f_i} b_{f_j} + U_{ij}^{--} b_{f_i} b_{f_j} + h.c. \right),$$

where couplings $V_{ij}^{\alpha\beta}$ and $U_{ij}^{\alpha\beta}$ are non–zero only for the nearest neighbours magnetic ions and they fulfil the following symmetry conditions:

$$V_{2ij}^{\alpha\beta} = V_{2i-1j}^{\alpha\beta},$$
$$V_{ij}^{\alpha\beta} = V_{ji}^{\beta\alpha} = V_{ij}^{-\alpha-\beta},$$
$$V_{13}^{++} = 0,$$
$$V_{13}^{+-} = -J\cos(2\alpha),$$
$$V_{15}^{+-} = V_{35}^{+-} = -\frac{J+K}{2}\cos(\alpha) - \frac{J-K}{2}\sin(\alpha),$$
$$V_{35}^{++} = -V_{15}^{++} = \frac{J-K}{2}\cos(\alpha) + \frac{J+K}{2}\sin(\alpha),$$
$$U_{15}^{\alpha\beta} = U_{35}^{\alpha\beta} = 0,$$
$$U_{13}^{+-} = U_{13}^{++} = K\sin^2(2\alpha).$$

The diagonalization of the Hamiltonian can be performed via a canonical transformation of bosonic operators [30] similarly to the previous work [18]. By this method the eigenvalues of two matrices 12×12 should be calculated. We have solved the problem in two steps. First, the elements of the matrices were calculated algebraically in Reduce and they were saved as Fortran subroutines for further numerical analysis which was performed as the second step.

The results of numerical calculation show that the energy excitations are positive for those values of anisotropy constants ($\kappa = K/J$, $d = D/2J$) which lie inside the wedge of fig. 3. Thus the negative excitation in U_3Sb_4 were obtained in earlier work [18] due to the application of standard spin wave description which is inadequate for this case.

The magnon dispersive curves presented in fig. 4 were obtained for the values $\kappa = 2.9$, $d = 5.1$ used earlier [10] to fit some experimental data for U_3Sb_4. As usual we analysed the dependence of energy excitation $\epsilon_k = E_k/2J$ on the wave vector k along some symmetry lines of the Brillouin zone. There are 12 branches, i.e., two times greater than in the standard spin wave approach [18]. The magnons of the b boson operators have a high energy excitation and a weak k–dependency. The lowest energy value is 7.57 for the k–independent branch which is 2–fold degenerated. The remaining branches of these types are located in the top of the diagram and are weakly k–dependent. On the other hand the branches corresponding to the a operators have a behaviour similar to the typical spin waves with degeneracy at special points of the Brillouin zone the same as obtained in the earlier work [18].

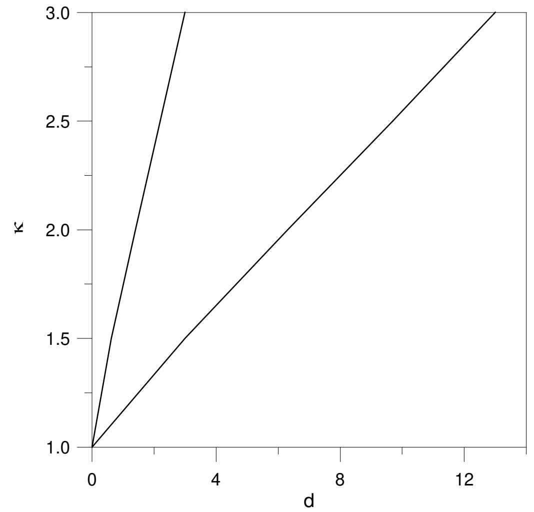

Fig. 3. Region of model parameters (inside the wedge) where energy excitations are positive.

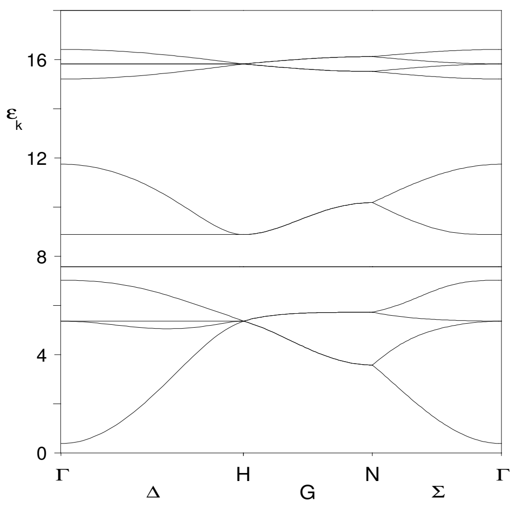

Fig. 4. Energy spectrum calculated for $\kappa = 2.9, d = 5.1$. The symmetry points of the Brillouin zone for the body centred cubic lattice: Γ=(0,0,0), H=$2\pi/a$(0,1,0), and N=π/a(1,1,0).

The presence of the energy gap in the spectrum (the lowest branch has non–zero value at the Γ point (see fig. 4)) is important for the temperature dependency of some quantities, e.g., magnetisation, heat capacity. The energy gap depends on the model parameters (d,κ) which is presented in fig. 5.

It is worth emphasizing that transformation (19) which diagonalizes the single–ion Hamiltonian (15) is the crucial step in the proper description of low temperature magnon excitations in the magnetic structure of U_3Sb_4. We checked that the application of the standard Holstein–Primakoff transformation to the transformed Hamiltonian leads to an effective harmonic magnon Hamiltonian the same as a part of $\mathcal{H}_{SWA}$ (39) depending on the operators a^+, a. In this way we obtained the same region of positive excitations as indicated in fig. 3 and the energy spectrum as the lower part of the spectrum presented in fig. 4.

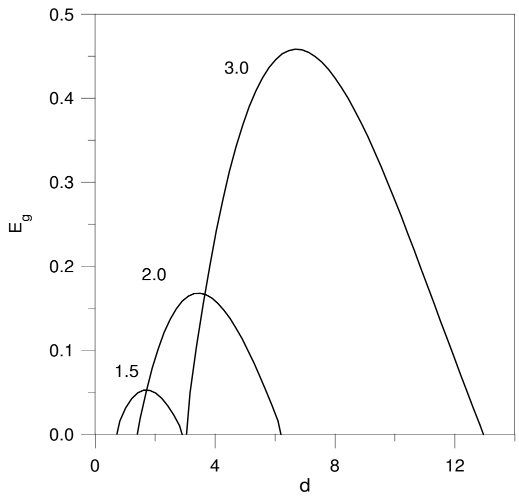

Fig. 5. Energy gap as a function of crystal field constant d for $\kappa = 1.5,\ 2,\ 3$.

VI. DISCUSSION

Using the mean field approximation at temperature $T = 0$ we demonstrated the role of the crystal field single–ion anisotropy. Its presence does not allow for the saturation of the sublattice magnetisation. The more drastic reduction of the magnetisation at $T = 0$ is obtained in the collinear phase of U_3Sb_4 than in the non–collinear magnetic structure of U_3P_4. We think this is the reason why the standard harmonic spin wave approach gives unphysical results for excitation in the collinear phase for any values of model parameters.

We demonstrated explicitly that introducing the quasi–particle idea as proposed by Onufrieva [21] does not require accounting for anharmonic terms in bosonic operators in order to describe low temperature excitation. The energy spectrum calculated by making use of this method for U_3Sb_4 (see fig. 4) has the energy gap that should affect the temperature dependence of magnetisation and heat capacity. The number of branches in the spetrum is 12, i.e., two times greater than in the standard spin wave approach. The additional branches weakly depend on the wave vector.

It would be interesting for the use the Onufrieva method to excitations in noncollinear magnetic structure eq. (8) in order to compare these with the earlier calculated spectra [18]. We expect only quantitative differences for small spin values and large crystal field anisotropy.

Finally, we want to discuss the influence of the relativistic anisotropy on the spectra for U_3Sb_4. It has been shown [29] by making use of the standard spin wave approach that adding to the Hamiltonian (4) the broken Dzialoshinski–Moriya term [16] changes the collinear structure (9) of U_3Sb_4 into that described by formula (3), i.e., a small perpendicular antiferromagnetic arrangement occurs. On the other hand this anisotropy removes the degeneracy of excitation at special points of the Brillouin zone and slightly changes the energy of excitations. As we know from the experimental mesuarement [11] the value of this anisotropy is rather very small with respect to the exchange interaction hence we suppose such a relativistic anisotropy would cause only small changes in the calculated magnon spectrum.

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СПІНОВІ ХВИЛІ В U_3Sb_4

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Використовуючи метод Онуфрієвої, проаналізовано спін–хвильові збудження в магнетних структурах U_3Sb_4. Запропонований аналіз усуває труднощі, пов'язані з від'ємними енергіями магнонів, відзначені в попередніх публікаціях. Виконано розрахунки для спіну $S = 1$ вздовж деяких ліній зони Бриллюена.