ELECTRON AND NUCLEAR SPIN CONFIGURATIONS AND INDUCED MAGNETOSTRICTION OF "EASY-PLANE" TRIANGULAR ANTIFERROMAGNETS IN THE EXTERNAL MAGNETIC FIELD

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We consider the magnetic and magnetostrictive properties of triangular antiferromagnets with the ABX_3 structure. At arbitrary ratios between the integrals of exchange in a plane and between the planes, the dependence of three turn angles of the magnetic moments of ions of the sublattices on the external magnetic field is determined. It is shown that, at any ratios between those parameters, there are two critical slamming fields. The nuclear spin configurations of such compounds are analyzed. The dependence of the exchange and single-ion rhombohedral magnetostrictions on the external field is studied. All calculations were made for the homogeneously ordered state.

1. Introduction

In the last years, the interest of researchers has noticeably shifted from collinear structures to noncollinear ones, especially if the noncollinearity has the exchange nature, rather than the relativistic one. (see e.g, [1—3]). In such magnets possessing usually a high spatial symmetry (the principal crystallographic axis C_n has rank n=3 or 6), the number of sublattices is more than two, and their description is complicated. In this case, the main questions are how, when, and under which conditions the number of sublattices can be diminished in order to reduce the description to well-elaborated two- and three-sublattice models. The latter model became especially actual due to the intense study of the family of double halogens with common chemical formula ABX_3 (among them, we mention $CsMnBr_3$, $CsNiCl_3$, $RbFeF_3$, $RbMnBr_3$, and others [1,2]), for which almost all experimental results are interpreted from the positions of the model with three magnetic sublattices in a nonstrained crystal.

We recall that the exchange-coupled spins of d-ions (Mn^{2+}, Fe^{3+}, etc.) in the above-mentioned compounds lie in the hexagonal planes. If the exchange interaction dominates among spin-spin ones and has the antiferromagnetic character, then the formation of a collinear (two-sublattice) structure turns out to be disadvantageous, and a noncollinear 120-degree (triangular) spin ordering becomes the most stable. Just this structure was proposed in [4] (see also [5]) for the β-phase of solid oxygen that has rhombohedral structure. Though the matter in both cases concerns crystals that are formed by hexagonal planes, halogenides have the C_6 axis, and β-O_2 has the C_3 axis. Consequences of the denoted distinction should be considered quite important, and they consist in the existence of the hierarchy of intra- and intersublattice interaction parameters. In particular, the interplane exchange field is absent in β-O_2 because of the rhombohedral structure. So, hexagonal planes become practically isolated from one another in the magnetic sense. In other words, β-O_2 is practically a 2D magnet, and the three-sublattice approximation that is adequate to this situation does not need any additional reasoning and is used in the framework of both the phenomenological and microscopic approaches [6—9] (see review [10]). Returning to ABX_3-type compounds, we note that the spins of B^{2+} magnetic ions form, on the one side,

antiferromagnetic chains along the crystallographic axis C_6 through the halogenide atoms which surround them and, on the other side, triangular structures in the basic plane [11]. At the same time, the exchange interactions in a plane and between planes, being antiferromagnetic, are directly opposite by their magnitude. The main interaction is the interplane one, whose "constant" is more than by two orders greater than that of the in-plane interaction. In this case, the considered structures are close to 1D magnets[1]. In addition, the number of sublattices in ABX_3 systems doubles (from 3 to 6). Therefore, the number of spin-wave branches grows, though only low-frequency excitations [13—16] can be successfully described in the framework of the microscopic approach developed in [12].

On the other side, even low-frequency excitations were studied till now without consecutive account of the influence of the nuclear spin subsystem and the magnetostriction on them which is specially strong in easy-plane magnets [17]. Moreover, both mentioned interactions in triangular antiferromagnets have a specificity related to the absence of nonisotropic striction in the initial 120-degree structures. At the same time, just the magnetostriction can stimulate phase transformations in them [18,19].

The above-said puts the questions analyzed in [20] to theory about the necessity of the consistent investigation of spin (electron and nuclear) excitations in the triangular antiferromagnets with "easy-plane" anisotropy. This investigation, as known, must be forestalled by the computation of spin configurations in the ground state of a crystal. Below, we give the solution of the latter problem with regard for their magnetostrictive features.

2. Hamiltonian and Spin Configurations

Intending to elucidate the magnetic-field characteristics of 6-sublattice triangular easy-plane spin systems, we write down their model Hamiltonian as

$$H = H_{1D} + H_{2D} + H_{an} + H_{STI} + H_Z, \tag{1}$$

where the terms

$$H_{1D} = I_{ch} \sum_{\mathbf{n}_\alpha, \rho_{\alpha\alpha}} \mathbf{S}_{\mathbf{n}_\alpha}(1)\, \mathbf{S}_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}}(2) \tag{2}$$

and

$$H_{2D} = I_{pl} \sum_{j=1,2} \sum_{\mathbf{n}_\alpha, \rho_{\alpha\beta}, (\beta\neq\alpha)} \mathbf{S}_{\mathbf{n}_\alpha}(j)\, \mathbf{S}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}}(j) \tag{3}$$

correspond to the chain (1D) and plane (2D) Heisenberg Hamiltonians with positive exchange (antiferromagnetic) constants I_{ch} and I_{pl};

$$H_{an} = -\Delta I_{ch} \sum_{\mathbf{n}_\alpha, \rho_{\alpha\alpha}} S^y_{\mathbf{n}_\alpha}(1)\, S^y_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}}(2) \tag{4}$$

— the operator of the anisotropy that was chosen in the simplest ionic form under the assumption that $\Delta I_{ch} > 0$[2] (in this case, the OY axis is chosen along the symmetry axis C_6);

$$H_{STI} = -A_0 \sum_{j=1,2} \sum_{\mathbf{n}_\alpha} \mathbf{S}_{\mathbf{n}_\alpha}(j)\, \mathbf{I}_{\mathbf{n}_\alpha}(j) \tag{5}$$

— the operator of the hyperfine interaction, where $A_0 > 0$ according to [17]; for simplicity, we consider such an interaction to be isotropic, and the parameters of the superhyperfine interaction are neglected (it is also assumed that nuclear spins for all atoms of this kind are the same, i.e. we do not consider the real isotope composition, when not all nuclei have intrinsic magnetic moment); at last,

$$H_Z = H_Z^{(e)} + H_Z^{(n)} =$$

$$= -\mathbf{H} \sum_{j=1,2} \sum_{\mathbf{n}_\alpha} [\mu_B g_\perp^{(e)} \mathbf{S}_{\mathbf{n}_\alpha}(j) + \mu_n g_\perp^{(n)} \mathbf{I}_{\mathbf{n}_\alpha}(j)] \tag{6}$$

— the operator of the Zeeman energy of electron $H_Z^{(e)}$ and nuclear $H_Z^{(n)}$ spins in a transverse magnetic field $\mathbf{H} \perp OY$ (everywhere further, it is considered that the magnetic field is perpendicular to the OY axis, i.e. $|\mathbf{H}| = H_\perp$).

In (2)—(6), we used the next designations: $\mathbf{S}_{\mathbf{n}_\alpha}(j)$ — the electron spin operator S in the cell $\mathbf{n}$ of the sublattice α (=1, 2, 3) from a plane of the j (j=1, 2) type; $\mathbf{I}_{\mathbf{n}_\alpha}(j)$ — the nuclear spin operator I; $\rho_{\alpha\beta}$ — the vector that connects the nearest spins from one ($\alpha = \beta$) and different ($\alpha \neq \beta$) magnetic sublattices (or, what is the same, planes, since the numeration in different

[1] Let us note that, in a good approximation, β-O_2 and and ABX_3 can be described, respectively, by two quasi-2D and three quasi-1D structures.

[2] Relation (4) is used more often with single-ion anisotropy, whose consideration requires to apply the quantum approach [21,22]. However, if it is small, which occurs in the Mn^{2+}- and Fe^{3+}-based halogenides, the results of the quasiclassic and quantum approaches coincide. It is natural to think that the constant ΔI_{ch} in (4) has the dipole-dipole origin.

planes repeats); $\mu_{\rm B}$ and μ_n — the electron and nuclear magnetons; and $g_{\perp}^{(e)}$ and $g_{\perp}^{(n)}$ — the corresponding g-factors. We note also that both the exchange and hyperfine constants are phenomenological parameters, rather than the calculated ones. Their values can be determined on the basis of the analysis of some experimental data which testify that, in the Mn^{2+}- and Fe^{3+}-based easy-plane systems, $I_{\rm ch} >> \Delta I_{\rm ch} >> I_{\rm pl}$ [2,20], the anisotropy $\Delta I_{\rm pl}$ is omitted. In β-O_2, however, the largest is the constant $I_{\rm pl}$. Therefore we will consider below that the ratio of $I_{\rm pl}$ and $I_{\rm ch}$ is arbitrary.

Then we will proceed to the proper coordinate systems $\xi\eta\zeta$, by considering that the O_{ζ_α} axis is the axis of spin quantization of each sublattice. The corresponding transition is described by the rotation angles $\varphi_\alpha(j)$ that are reckoned from the field direction (see Fig. 1):

$$\left\{\begin{array}{l} S^X_{\mathbf{n}_\alpha}(j) = S^\xi_{\mathbf{n}_\alpha}(j)\cos\varphi_\alpha(j) \pm (-1)^j S^\zeta_{\mathbf{n}_\alpha}\sin\varphi_\alpha(j), \\ S^Y_{\mathbf{n}_\alpha}(j) = S^\eta_{\mathbf{n}_\alpha}(j), \\ S^Z_{\mathbf{n}_\alpha}(j) = \mp(-1)^j S^\xi_{\mathbf{n}_\alpha}(j)\sin\varphi_\alpha(j) + S^\zeta_{\mathbf{n}_\alpha}\cos\varphi_\alpha, \end{array}\right. \tag{7}$$

where upper signs in the combinations $\pm$ and $\mp$ occur at $\alpha = 1$, and lower signs at $\alpha = 2,3$. The operators $\mathbf{I}_{\mathbf{n}_\alpha}(j)$ are transformed similarly. In this case, it should be born in mind that, as usual, the constant $A_0 >> \mu_n g_m H_\perp$, which yields that the knowledge of the directions of electron spins defines, in fact, the directions of the corresponding nuclear spins that are at the same sites. However, in order that the computations have the general character, we assume that the strong inequality is absent. So, we will determine the angles that correspond to nuclear spin moments that undergo the influence of both the hyperfine and external fields.

The further cumbersome, but simple computations are related to the use of transformation (7) of operator (1), in which we single out the terms linear in the operators $S^{\xi,\eta}_{\mathbf{n}_\alpha}(j)$ and $I^{\xi,\eta}_{\mathbf{n}_\alpha}(j)$ of electron and nuclear spins and equate them to zero. In this case, we get six equations in each case that minimize the ground-state energy in the quasiclassical approximation. By virtue of the evident symmetry of the problem, the spin directions in planes of types 1 and 2 are opposite. Therefore, it is sufficient to introduce only three "electron" angles and three "nuclear" ones. Taking the choice of the proper axes for spins in expressions (7) into account, these three angles for electron spins are as follows (see Fig. 1):

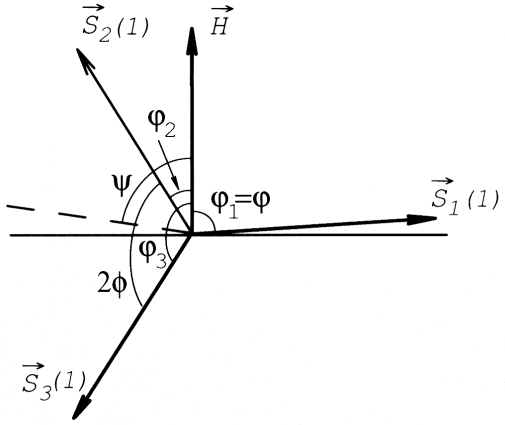

Fig. 1. Direction of magnetic sublattices in plane *1*. Plane *2* corresponds to both the turn of three vectors $\mathbf{S}_1(1)$, $\mathbf{S}_2(1)$, and $\mathbf{S}_3(1)$ by an angle of π in the plane and the interchange of $\mathbf{S}_3(2)$ and $\mathbf{S}_2(2)$. The dashed line shows the direction of the "summary" spin of the 2nd and 3rd sublattices: $\mathbf{S}_2(1) + \mathbf{S}_3(1)$

$$\varphi_1(1) = \varphi_1(2) = \varphi, \quad \varphi_2(j) + \varphi_3(j) = 2\psi,$$

$$\varphi_2(j) - \varphi_3(j) = (-1)^j 2\phi, \tag{8}$$

and, for nuclear spins,

$$\varphi_1^{(n)}(1) = \varphi_1^{(n)}(2) = \varphi^{(n)}, \varphi_2^{(n)}(j) + \varphi_3^{(n)}(j) = 2\psi^{(n)},$$

$$\varphi_2^{(n)}(j) - \varphi_3^{(n)}(j) = (-1)^j 2\phi^{(n)}. \tag{9}$$

As a result, we arrive at the next equations of equilibrium:

$$\left\{\begin{array}{l} I_{\rm ch} s z_{\rm ch}\sin 2\varphi + 2I_{\rm pl} s z_{\rm pl}\sin(\varphi+\psi)\cos\phi - \\ -\mu_{\rm B} g_\perp^{(e)} H_\perp \sin\varphi = 0, \\ I_{\rm ch} s z_{\rm ch}\sin 2\psi + I_{\rm pl} s z_{\rm pl}\sin(\varphi+\psi)\cos\phi - \\ -\mu_{\rm B} g_\perp^{(e)} H_\perp \sin\psi\cos\phi = 0, \\ I_{\rm pl} s z_{\rm pl}[\cos(\varphi+\psi)\sin\phi + \sin 2\phi] - \\ -\mu_{\rm B} g_\perp^{(e)} H_\perp \cos\psi\sin\phi = 0, \end{array}\right. \tag{10}$$

where $z_{\rm ch} = 2$ and $z_{\rm pl} = 3$ are, respectively, the numbers of the nearest neighbors from other sublattices in chains and planes. Similarly for nuclear spins, we get the equations

$$\left\{\begin{array}{l} H_n^{(0)}\sin(\varphi - \varphi^{(n)}) - H_\perp \sin\varphi^{(n)} = 0, \\ H_n^{(0)}\sin(\psi - \psi^{(n)})\cos(\phi - \phi^{(n)}) - \\ -H_\perp \sin\psi^{(n)}\cos\phi^{(n)} = 0, \\ H_n^{(0)}\sin(\psi - \psi^{(n)})\cos(\phi - \phi^{(n)}) - \\ -H_\perp \cos\psi^{(n)}\sin\phi^{(n)} = 0 \end{array}\right. \tag{11}$$

that just define the configurations of the corresponding spins in a transverse field. In the system of equations (10) and (11), we neglected the influence of nuclear spins on the directions of electron spins, used the standard notation $\mu_n g_\perp^{(n)} H_n^{(0)} \equiv A_0 s$ for the initial hyperfine field $H_n^{(0)}$ at a nucleus, and introduced the mean value of the electron spin $s = \left\langle S_{n_\alpha}^{\zeta}(j) \right\rangle$ at the node. In addition, we note that, according to [20], the relative spin reduction or the value $(S-s)/S$ that is conditioned by low-dimension effects in $CsMnBr_3$ reaches 30%. Therefore, the computation of the mean spin s should be realized not in the mean field approximation, but in the Tyablikov approximation [23].

The solution of system (11) follows from the known solution of system (10), for which, as is easy to verify, the most stable at $H_\perp \to 0$ is the structure $\varphi = \psi = \pi/2$, $\phi = 2\pi/3$, i.e. a 120-degree structure sometimes called the Loktev structure. According to (5) and (11), $\varphi^{(n)} = \psi^{(n)} = \pi/2$, $\phi^{(n)} = 2\pi/3$. In other words, nuclear spins become polarized and collinear to their "own" electron spins. Moreover, the magnetic structure that is realized in the absence of the external field and corresponds to Hamiltonian (1) is a multisublattice antiferrodielectric with "easy-plane" anisotropy.

If $H_\perp \neq 0$, then the solution of system (10) depends on the ratio $I_{\rm pl}/I_{\rm ch}$ and is the unique one. If we restrict ourselves by the noninteracting planes with three sublattices, i.e. the system is two-dimensional, then the number of the unknowns is larger than the number of equations, and the solution becomes ambiguous. In view of this fact, Stefanovsky [24] made conclusion that the three-sublattice isotropic two-dimensional antiferromagnets with easy-plane anisotropy placed in the external field that also lies in the plane reveal basically the continuous degeneracy or the continuum of equal-energy states. We note that the equations in [24] follow from (10) if $I_{\rm ch} = 0$. Such a degeneracy, even being mathematically correct, is hardly justified physically for real systems, because the account of the three-dimensional interplane coupling (in the considered case, it's $I_{\rm ch} \neq 0$) removes the mentioned degeneracy. Thus, all directions of spins of the magnetic sublattices are fully defined.

Formally, system (10) can correspond both to the case of ABX_3 where $I_{\rm ch} >> I_{\rm pl}$ and to β-O_2 if $I_{\rm ch} << I_{\rm pl}$. In the first case, there is an analytical solution of Eqs. (10) that was found in [12] for fields $H_\perp << H_{\rm s-f} \equiv 2H_{\rm ch}^{(E)} + 3H_{\rm pl}^{(E)}$, where $H_{\rm s-f}$ is the spin-flip field, $H_{\rm ch,pl}^{(E)} = I_{\rm ch,pl} s z_{\rm ch,pl}/\mu_{\rm B} g_\perp^{(e)}$ are the interplane and intraplane exchange fields. Since $H_{\rm ch}^E >> H_{\rm pl}^E$, the matter concerns, in fact, the inequality $H_\perp << H_{\rm ch}^{(E)}$ on the estimation $H_{\rm s-f} \approx 2H_{\rm ch}^{(E)}$. In this case, the relative values of the fields $H_\perp$ and $H_{\rm pl}^E$ remains arbitrary. Then, considering that $\varphi < \pi/2$ and $\psi \lesssim \pi/2$, we obtained from (10) to within terms $\sim (H_\perp/H_{\rm s-f})^2$ inclusively:

$$\cos\varphi = \frac{H_\perp}{2H_{\rm ch}^{(E)} + 3H_{\rm pl}^{(E)}\cos\phi} \approx \frac{H_\perp}{H_{\rm s-f}},$$

$$\sin\varphi = 1 - \frac{1}{2}(H_\perp/H_{\rm s-f})^2,$$

$$\cos\psi \equiv \begin{cases} \cos\varphi\cos\phi, & H_\perp \le H_{\rm cr}, \\ \cos\varphi, & H_\perp > H_{\rm cr}; \end{cases}$$

$$\sin\psi \equiv \begin{cases} 1 - \frac{1}{2}(H_\perp/H_{\rm s-f})^2\cos^2\phi, & H_\perp \le H_{\rm cr}, \\ \sin\varphi, & H_\perp > H_{\rm cr}. \end{cases} \tag{12}$$

Here, the critical field $H_{\rm cr} = \sqrt{2H_{\rm ch}^{(E)} H_{\rm pl}^{(E)}}$ [12], according to the relation

$$\cos\phi \equiv \begin{cases} [2 - (H/H_{\rm cr})^2]^{-1}, & H_\perp \le H_{\rm cr}, \\ 1, & H_\perp > H_{\rm cr} \end{cases} \tag{13}$$

defines the behaviour of the angle ϕ in all orders of the ratio $H_\perp/H_{\rm cr}$. It is obvious that this angle, by changing from $2\pi/3$ to 0, defines (see Fig. 1) the counterrotation of two (of three) antiferromagnetic vectors $\mathbf{l}_\alpha = \mathbf{S}_{\mathbf{n}_\alpha}(1) - \mathbf{S}_{\mathbf{n}_\alpha}(2)$ that correspond to $\alpha = 2$ or 3, to the most favorable energy state, when $\mathbf{l}_\alpha \perp \mathbf{H}_\perp$ (we note that, according to the accepted sublattice numbering, $\mathbf{l}_1 \perp \mathbf{H}_\perp$ by definition).

As the ratio $I_{\rm pl}/I_{\rm ch} = a$ grows, expressions (12) and (13) are no longer enough precise solutions, and system (10) was analyzed numerically.

One of the results of such numerical analysis is the dependence of the angles of the sublattice spin directions on the external field that is shown on Fig. 2. All calculations here and further were made, for certainty, at some "intermediate" value of the parameter, $a = 0.3$. It turns out that the topology of the curves obtained has no significant distinctions from that shown on Fig. 2, neither at $a >> 1$, nor at $a << 1$. The values of critical fields are different, and the breaks at critical points are manifested differently, but always there occurs, at first, the "cohesion" of two of three sublattices in each plane. Then, in the spin-flip field, the multisublattice magnet is

transformed into a one-sublattice one. Thus, the angle ϕ shows the simplest behaviour. At the point $H_{\rm cr}$, it reaches zero (sublattices 2 and 3 slam) and then is not changed. But the angles φ and ψ have a break in the first critical field, change the character of their dependence, and reach the zero value in the field $H_{\rm s-f}$ (the transition to the ferromagnetic state).

3. Nuclear Spin Configurations

The average nuclear spin orientations (for the nuclei of ions that belong to one or other sublattice), i.e. the solution of the system of equations (11), can be easily found precisely by the known electron spin configuration. We will write down this solution in the approximation used in this work (i.e. $\varphi^{(n)} \le \pi/2$, $\psi^{(n)} \le \pi/2$). In this case, as was mentioned earlier, $\varphi^{(n)} \neq \varphi$ and $\psi^{(n)} \neq \psi$, because nuclear spins are, generally speaking, under the influence of two fields, the external and hyperfine ones[3]. The sought expression can be easily obtained under the condition that $H_\perp << H_n^{(0)}$ that agrees with the observed values (see [19], where the angles are found without regard for the terms $\sim H_\perp/H_{\rm s-f}$). They have the form

$$\cos\varphi^{(n)} = \frac{H_\perp}{H_n^{(0)}} + \frac{H_\perp}{H_{\rm s-f}},$$

$$\sin\varphi^{(n)} = 1 - \frac{1}{2}\left(\frac{H_\perp}{H_n^{(0)}} + \frac{H_\perp}{H_{\rm s-f}}\right)^2,$$

$$\cos\psi^{(n)} = \cos\varphi^{(n)},$$

$$\sin\psi^{(n)} = 1 - \frac{1}{2}\left(\frac{H_\perp}{H_n^{(0)}} + \frac{H_\perp}{H_{\rm s-f}}\right)^2 \cos^2\phi,$$

$$\cos\phi^{(n)} = \cos\phi + \left(\frac{H_\perp}{H_n^{(0)}}\right)^2 \sin^2\phi\cos\phi +$$

$$+\frac{H_\perp^2}{H_n^{(0)} H_{\rm s-f}} \sin^2\phi,$$

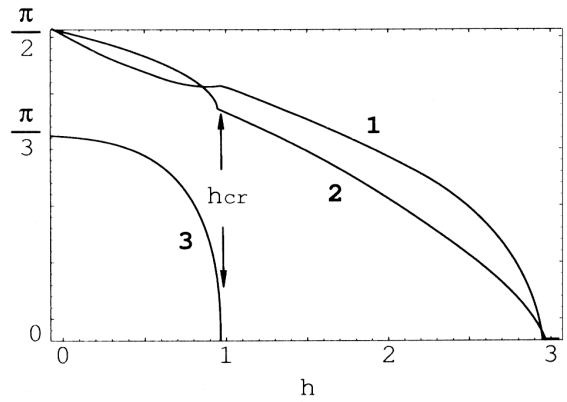

Fig. 2. Dependence of the generalized angles on the reduced magnetic field $h = H_\perp/H_{\rm ch}^{(E)}$ at $a = 0.3$. Curve *1* describes the behaviour of the angle ψ, *2* — φ, *3* – ϕ. Here and later on the figures, $h_{\rm cr} = H_{\rm cr}/H_{\rm ch}^{(E)}$

$$\sin\phi^{(n)} = \sin\phi - \left(\frac{H_\perp}{H_n^{(0)}}\right)^2 \sin\phi\cos^2\phi -$$

$$-\frac{H_\perp^2}{H_n^{(0)} H_{\rm s-f}} \sin\phi\cos\phi. \qquad (14)$$

Moreover, by virtue of $H_n^{(0)} >> H_{\rm cr}$, relations (14) are correct in the range $0 \le H_\perp \le H_n^{(0)}$.

In fact, expressions (14) [together with (12), (13)] exhaust the problem of spin (electron and nuclear) configurations that appear in easy-plane triangular antiferromagnets of the ABX_3 type. In these systems, because of the strong inequality $I_{\rm pl} << I_{\rm ch}$, the solution of system (10) is well described by the known relations for two-sublattice antiferromagnets in a transverse ($\mathbf{H}_\perp \perp OY$) field after the first (in the field $H_\perp = H_{\rm cr}$) spin-flip, when $\phi \to 0$: $\cos\varphi = \cos\psi = H_\perp/2H_{\rm s-f} \simeq H_\perp/2H_{\rm ch}^{(E)}$. Though the role of the exchange interaction $I_{\rm pl}$ consists only in the formation of a finite range of the swivel of antiferromagnetic vectors $\mathbf{l}_\alpha$ (at $\alpha = 2.3$), it should be born in mind that the field $H_{\rm cr}$ (see (13)) do not make all sublattices equivalent. In fact, a skewed ferrimagnetic structure with the "short" s and "long" $2s$ spins is formed in each sheet (see Fig. 1), where the corresponding "sublattices" cannot be considered equivalent in the physical sense. The last means that $\varphi \neq \psi$ in such a structure in strict sense, although the difference can be relatively small.

[3]It should be mentioned that only the nuclei of nonmagnetic ions can have nonzero spins (in the case of an ABX_3-type system, it can concern the elements A and X). Then their interaction with electron spins of transition metals will be nonlocal, and the corresponding constant of a superhyperfine bond is essentially less than A_0. In this case, the formulas that are obtained below and define the directions of nuclear spins can implicitly describe nuclear spin configurations of nonmagnetic ions, although such a problem needs a separate analysis on the whole.

4. Magnetostriction of Triangular Magnets

As was mentioned above, the influence of the magnetoelastic interaction has not been taken into account so far in the works considering the properties and spin-wave excitation in easy-plane antiferromagnets of the ABX_3 type. This section is devoted to the analysis of its consequences, first of all, for magnetostriction.

The consideration of magnetostriction effects can be divided into two parts which corresponds to the types of the interaction that will be taken into account. First, we will account only the exchange interaction and the exchange magnetostriction induced by it. In the framework of such a problem, there can be used the same quasiclassical approximation as in the consideration of the spin configuration problem, and the single-ion anisotropy can be neglected. In this case, the Hamiltonian that describes such an interaction can be written as

$$\widetilde{H} = \widetilde{H}_{1D} + \widetilde{H}_{2D} + H_Z + \Phi, \tag{15}$$

where the terms

$$\widetilde{H}_{1D} = \sum_{\mathbf{n}_\alpha, \rho_{\alpha\alpha}} I_{ch}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}}) \mathbf{S}_{\mathbf{n}_\alpha}(1)\, \mathbf{S}_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}}(2) =$$

$$= H_{1D} + H^{str}_{exch}(ch) \tag{16}$$

and

$$\widetilde{H}_{2D} = \sum_{j=1,2} \sum_{\mathbf{n}_\alpha, \rho_{\alpha\beta}} I_{pl}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}}) \mathbf{S}_{\mathbf{n}_\alpha}(j) \times$$

$$\times \mathbf{S}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}}(j) = H_{2D} + H^{str}_{exch}(pl) \tag{17}$$

correspond, like in the previous model, to the chain and plane Heisenberg magnets with the exchange integrals $I_{ch}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}})$ and $I_{pl}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}})$, and H_{1D} and H_{2D} are set by expressions (2) and (3). Here, we took into account that, as distinct from the previous case, the integrals are not constants, but depend on the arrangement of atoms in the lattice. Here, $\mathbf{R}_{\mathbf{n}_\alpha}$ is the position of atoms in the lattice after the deformation; $H_Z \equiv H_Z^{(e)}$ corresponds to the Zeeman Hamiltonian of electron spins in the magnetic field that is transverse to the anisotropy axis;

$$\Phi = Nv\{\frac{K}{2}(u_{xx}+u_{zz})^2 + \frac{\mu}{2}\left[(u_{xx}-u_{zz})^2 + 4u_{xz}^2\right] +$$

$$+\frac{1}{2}c_1 u_{yy}^2 + 2c_2 u_{yy}(u_{xx}+u_{zz})\} \tag{18}$$

is the elastic part of the energy that is related to a deformation of the system (see [25]), where N is the number of spins; v is the volume per spin; u_{ij} are components of the strain tensor; K, μ, c_1, and c_2 are the corresponding moduli of elasticity. The formula for such an energy is defined by a symmetry of the considered lattice. In our case, the crystal has hexagonal symmetry with the axis $C_6 \parallel Y$, i.e. the basic plane is isotropic.

We expand the exchange integrals $I_{ch}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\alpha}})$ и $I_{pl}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}})$ in the shifts of atoms from the equilibrium positions and account only the terms that are linear in deformation:

$$I_{pl,ch}(\mathbf{R}_{\mathbf{n}_\alpha}, \mathbf{R}_{\mathbf{n}_\alpha+\rho_{\alpha\beta}}) = I_{pl,ch}(1 - \lambda^{(pl,ch)}_{exch} e^i_\rho u_{ij} e^j_\rho). \tag{19}$$

Here, $\lambda^{(pl,ch)}_{exch} = -a_{pl,ch}\partial \ln I_{pl,ch}/\partial\rho$ is the constant of the magnetoelastic interaction of the exchange nature, $a_{pl,ch}$ is the constant of the undistorted lattice in a plane and between planes, ρ=(1, ... ,8) numbers the nearest neighbors of a given atom, $a_{pl,ch}e^i_\rho$ $(i = x, y, z)$ is the Cartesian projection of the vector that connects the given site with its nearest neighbor. As a result, we easily comes to the expressions for H^{str}_{exch}(ch) and H^{str}_{exch}(pl) which are not given here.

Further, like in (7) and (8), we pass to the proper coordinate systems for each spin sublattice in all terms of Hamiltonian (15). Like above, we reject all terms that are proportional to the operators $S^\xi_{\mathbf{n}_\alpha}(j)$ and $S^\eta_{\mathbf{n}_\alpha}(j)$, leaving only the terms with products of $S^\zeta_{\mathbf{n}_\alpha}(j)$, because just they make the main contribution to the energy, and will consider $S^\zeta_{\mathbf{n}_\alpha}(j)$ to be numbers, rather than operators: $\langle S^\zeta_{\mathbf{n}_\alpha}(j)\rangle \approx s$.

Now we will find the magnitudes of various deformations in the crystal and their dependences on the external magnetic field from the minimum condition for Hamiltonian (15) relative to these deformations at the given values of the field. As a result, we get

$$u_{xx} + u_{zz} = \frac{s^2}{4c_2^2 v - Kc_1 v}\big\{4c_2\lambda^{(ch)}_{exch} I_{ch}\big(\cos 2\varphi +$$

$$+2\cos 2\psi\big) - 6c_1\lambda^{(pl)}_{exch} I_{pl}\big(\cos(\varphi+\psi-\phi) +$$

$$+\cos(\varphi+\psi+\phi\big) + \cos 2\phi)\big\}, \tag{20}$$

$$u_{yy} = \frac{s^2}{c_1 v}\big\{2\lambda^{(ch)}_{exch} I_{ch}(\cos 2\varphi + 2\cos 2\psi) -$$

$$-\frac{8c_2}{4c_2^2 v - KC_1 v}\big[c_2\lambda^{(ch)}_{exch} I_{ch}(\cos 2\varphi + 2\cos 2\psi) -$$

$$-3c_1\lambda^{(pl)}_{exch} I_{pl}(\cos(\varphi+\psi)\cos\phi + \cos 2\phi)\big]\big\}, \tag{21}$$

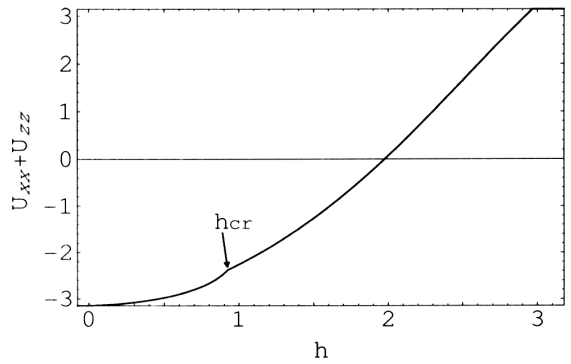

Fig. 3. Dependence of the reduced deformation $U_{xx} + U_{zz} = (u_{xx} + u_{zz})\frac{Kc_1v}{4s^2c_2\lambda^{(\mathrm{ch})}_{\mathrm{exch}}I_{\mathrm{ch}}}$ on the external magnetic field, when condition (23) is satisfied

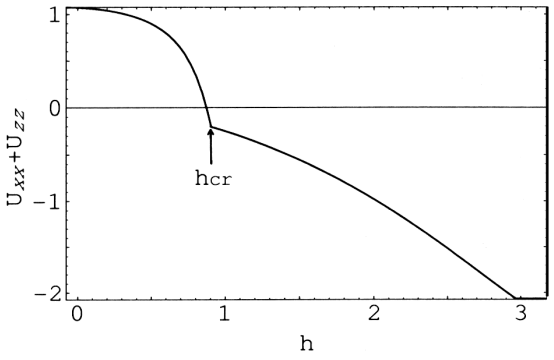

Fig. 4. Dependence of the reduced deformation $U_{xx}+U_{zz} = (u_{xx}+u_{zz})\frac{Kv}{6s^2\lambda^{(\mathrm{pl})}_{\mathrm{exch}}I_{\mathrm{pl}}}$ on the external magnetic field, when condition (25) is satisfied

$$u_{xx} - u_{zz} = u_{xz} = 0. \tag{22}$$

In this case, the directions of spins (the generalized angles φ, ψ, and ϕ) should be determined, generally saying, again from the minimum condition for the full Hamiltonian (15), by substituting values of the deformations according to (20)–(22). But, for simplicity, we will consider these angles to be those determined in Section 2, by neglecting small effects related to their variations at the expense of the magnetostriction (due to its relative smallness). Thus, under the consideration of only the exchange component of the magnetostriction for the examined class of magnets, only two diagonal components of the strain tensor, $u_{xx} + u_{zz}$ and u_{yy}, are nonzero. In other words, in the framework of the exchange approximation, the symmetry of the system will not change even under the switching-on of the external field of an arbitrary value.

It is evident from (20) and (21) that the deformation depends not only on $H_\perp$ and the ratio of I_{pl} and I_{ch}, but essentially on the ratios between K, μ, c_1, and c_2. Some variants of such ratios will be considered in what follows.

It was indicated earlier that the interplane exchange interaction in compounds of the ABX_3 type is essentially larger than the intraplane interaction. It will also be considered that the more the exchange integral, the more its derivative with respect to shifts (though this assumption cannot valid in the general case). On the other hand, those moduli of elasticity are considered to be greater that correspond to more symmetric terms. Therefore, the modulus of elasticity c_2 must be lower than c_1, at first sight. In this situation, we can consider two extreme cases.

1-st case:

$$4c_2\lambda^{(\mathrm{ch})}_{\mathrm{exch}}I_{ch} > 6c_1\lambda^{(\mathrm{pl})}_{\mathrm{exch}}I_{\mathrm{pl}}. \tag{23}$$

Then the expressions for the deformation will take the next form:

$$u_{xx} + u_{zz} = -\frac{4s^2}{Kc_1v}c_2\lambda^{(\mathrm{ch})}_{\mathrm{exch}}I_{\mathrm{ch}}(\cos 2\varphi + 2\cos 2\psi),$$

$$u_{yy} = \frac{2s^2}{c_1v}\lambda^{(\mathrm{ch})}_{\mathrm{exch}}I_{\mathrm{ch}}(\cos 2\varphi + 2\cos 2\psi).$$

The typical example of the dependence of the value $u_{xx} + u_{yy}$ on the reduced magnetic field is shown on Fig. 3. As was mentioned earlier, all computations are made here and below for some intermediate magnitude of the parameter a, which allows us to trace the features that correspond to large values of a (this is inherent to β-O_2) and to smaller ones (this corresponds to systems ABX_3).

2-nd case:

$$4c_2\lambda^{(\mathrm{ch})}_{\mathrm{exch}}I_{\mathrm{ch}} < 6c_1\lambda^{(\mathrm{pl})}_{\mathrm{exch}}I_{\mathrm{pl}}. \tag{24}$$

Then, for example, the expression for $u_{xx} + u_{yy}$ takes the form

$$u_{xx} + u_{zz} = \frac{6s^2}{Kv}\lambda^{(\mathrm{pl})}_{\mathrm{exch}}I_{\mathrm{pl}}\big\{\cos(\varphi + \psi - \phi) +$$

$$+\cos(\varphi + \psi + \phi) + \cos 2\phi\big\}. \tag{25}$$

The corresponding dependence on the external magnetic field is shown in Fig. 4.

Evidently, in the first case, the deformation in the easy magnetization plane at $H_\perp \to 0$ is negative (at the

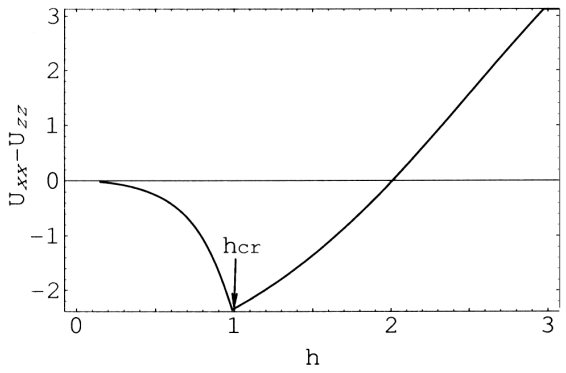

Fig. 5. Dependence of the reduced deformation (distortion, yield) $U_{xx}-U_{zz}=\frac{v\mu}{2s^2E}(u_{xx}-u_{zz})$ on the external magnetic field on the action of the relativistic magnetostriction

same signs of the constants c_1 and c_2). With increase of the field, it grows and becomes positive. In the second case, for the same values of the parameters, the situation turns out to be opposite. In both considered cases, irrespective of the ratios between the exchange constants and the moduli of elasticity, there appear certain peculiarities near critical fields. In the neighborhood of the spin-flip field $h_{\mathrm{s-f}}$, the deformation reaches the maximum. Then it stop to change with increase of the external field. In the field h_{cr}, we observe a break on the curve of the deformation versus the field.

As was noted, two types of the interaction lead to the appearance of magnetostriction. The one considered above corresponds to magnetostriction of the exchange type that does not lead to a change of the lattice symmetry in triangular antiferromagnets. Let us pass now to the analysis of the relativistic magnetostriction that just leads to a change of the lattice symmetry.

To this end, we will introduce the additional term $H_{\mathrm{rel}}^{\mathrm{str}}$ into Hamiltonian (15) of the system. On the phenomenological level, this term takes into account that such an interaction influences only the deformation in the basic plane XZ:

$$H_{\mathrm{rel}}^{\mathrm{str}}=E(u_{xx}-u_{zz})\sum_{\mathbf{n}_\alpha,j}\left\{\left(S_{\mathbf{n}_\alpha}^{x}(j)\right)^2-\left(S_{\mathbf{n}_\alpha}^{z}(j)\right)^2\right\}. \tag{26}$$

Here, E is the constant of the relativistic magnetostriction conditioned by the spin-orbit and dipole-dipole couplings. We can also consider that operator (27), if we say about the magnets with weak anisotropy [21], takes into account the contribution of the exchange interaction anisotropy.

Like in the previous case, we pass in the proper coordinate systems according to transformations (7) in (27) for all the operators $\mathbf{S}_{\mathbf{n}_\alpha}$. In this case, we will again omit the quantities that are proportional to $S_{\mathbf{n}_\alpha}^{\xi}(j)$ and $S_{\mathbf{n}_\alpha}^{\eta}(j)$, and replace $[S_{\mathbf{n}_\alpha}^{\zeta}(j)]^2$ by their numerical values, $(S_{\mathbf{n}_\alpha}^{\zeta}(j))^2\approx s^2$ (i.e. we neglect the difference between $\langle S_\zeta\rangle^2$ and $\langle S_\zeta^2\rangle$ that is small for weak-anisotropy systems [21]).

The dependence of the rhombic deformation of the lattice induced by a magnetic field on the field can be determined from the minimum condition for the ground-state energy of Hamiltonian (15) and (27) with respect to this deformation. As a result, by neglecting a small spontaneous magnetostriction [4], we get

$$u_{xx}-u_{zz}=-2\frac{E}{v\mu}s^2(\cos 2\varphi+$$

$$+\cos 2(\psi+\phi)+\cos 2(\psi-\phi)). \tag{27}$$

Thus, the account of the relativistic magnetoelastic interaction leads to the appearance of a rhombic deformation. However, the magnitude of such a deformation in crystals with the Jahn—Teller effect not too brightly evident turns out, generally, relatively small in comparison with other types of the deformation.

In Fig. 5, we present the characteristic dependence of the induced deformation (28) on the external magnetic field at the same magnitude of the parameter $a=0.3$.

Evidently, if we use system (10), then the deformation is absent at $H_\perp=0$. After the switching-on of the field, the system begins to elongate (for $E>0$) along one of its axis, and this extension reaches the maximum in the field h_{cr}. Under a further increase of the external field, the deformation start to decrease. At some field, it gets over zero, and henceforward it begins to grow along the second axis. In the spin-flip field, the system reaches the second maximum, and the further increase of the field do not change the deformation of the system. We note that, at an arbitrary value of the parameter a, the system has two phase transitions, and the magnetostriction will follow the dependence shown in Fig. 5. However, the position and the depth of the minimum can be changed. In any case, the rhombic distortion must affect the form of the spectrum of spin excitations.

[4]Its possibility indicated by S.M. Ryabchenko needs, however, an additional study.

5. Conclusion

We have performed the theoretical analysis of the magnetic features of triangular antiferromagnets, particularly, of compounds of the ABX_3 type and of their behaviour in the external magnetic field. The consideration was taken with regard for the Heisenberg exchange interaction for arbitrary ratios between the exchange integrals in a plane and between planes.

The obtained system of equilibrium equations allowed us to get the directions of spin sublattices in a magnetic field for each plane. It is shown that, for all ratios between the intra- and interplane exchange integrals, there exist two critical values of the field. We have described and analyzed the dependence of the nuclear-spin configurations on the external magnetic field.

A special attention is given to the study of the magnetostriction at arbitrary values of the external magnetic field. In this case, we have considered the isotropic deformation conditioned by the exchange and the induced rhombic distortion of the lattice that is related to single-ion relativistic interactions. In the numerical analysis of the magnetostriction, we considered that its back influence on the rotation angles of the magnetic sublattices can be neglected.

We nave demonstrated that the exchange magnetostriction in triangular structures leads to a change of the lattice parameters which is isotropic in a plane. In this case, the dependence of the deformation on the external field has monotonous character.

The analysis has also shown that a deformation leads to the field-induced elongation of the lattice along or across the field direction.

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ЕЛЕКТРОННІ І ЯДЕРНІ СПІНОВІ КОНФІГУРАЦІЇ ТА ВИМУШЕНА МАГНІТОСТРИКЦІЯ ТРІАНГУЛЯРНИХ АНТИФЕРОМАГНЕТИКІВ З АНІЗОТРОПІЄЮ ТИПУ "ЛЕГКА ПЛОЩИНА" У ЗОВНІШНЬОМУ МАГНІТНОМУ ПОЛІ

І.М. Іванова, В.М. Локтєв

Р е з ю м е

Розглянуто магнітні та магнітострикційні властивості тріангулярних антиферомагнетиків зі структурою ABX_3. Знайдено

залежність трьох кутів повороту магнітних моментів іонів підґраток від зовнішнього магнітного поля при довільних співвідношеннях між обмінними інтегралами у площині та між площинами. Показано, що для довільних співвідношень між вказаними параметрами існують два критичних значення поля схлопування напрямків намагніченості підґраток. Проаналізовано характер ядерних спінових конфігурацій таких з'єднань. Вивчено залежність обмінної та одноіонної ромбоедричної магнітострикцій від зовнішнього магнітного поля. Усі розрахунки проведено для однорідно впорядкованого стану.