Modification of the classical Heisenberg helimagnet by weak uniaxial anisotropy and magnetic field

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A classical ground state of the isotropic Heisenberg spin Hamiltonian on a primitive Bravais lattice is known to be a single-**Q** plane helix. Additional uniaxial anisotropy and external magnetic field can greatly distort this structure by generating higher-order (at the wave vectors $n\mathbf{Q}$) Fourier harmonics in the spatial spin configuration. These features are not captured within the usual formalism based on the Luttinger–Tisza theorem, when the classical ground state energy is minimized under the "weak" condition on the lengths of the spins. We discuss why the correct solution is lost in that approach and present another microscopic treatment of the problem. For easy-axis and easy-plane quadratic uniaxial anisotropy it allows one to find the classical ground state for general **Q** and for any orientation of the magnetic field considering the effect of anisotropy (but not the field) as a perturbation of the exchange structure. As a result, the classical ground state energy, the uniform magnetization, and the magnetic Bragg peak intensities that are measured in the experiments are calculated.

1. INTRODUCTION

For more than three decades, helical spin structures have been a subject of intensive studies. Having been discovered theoretically in the pioneering works of Yoshimori[1] and Villain,[2] they were found experimentally in a large variety of materials. Most extensive work was devoted to the investigation and explanation of the different ordered phases and phase transitions in the rare-earth metals from Tb to Tm. These phases were shown to result from the intricate distortion of the incommensurate, almost ferromagnetic exchange spiral by temperature and strong crystal field anisotropies.[3,4] However, for the reason discussed below, a classical treatment, finally generalized by Lyons and Kaplan,[5] predicts no such distortion in the ground state (g.s.) of the quasi-isotropic Heisenberg Hamiltonians at $T=0$.

A simple nontrivial example of the commensurate spiral is the triangular magnetic ordering found in the hexagonal antiferromagnets of $CsNiCl_3$ type. Their magnetic structure consists of six sublattices, three in each hexagonal plane at an angle $\approx 120°$ to each other, where the spins in the adjacent planes are antiparallel. Such compounds have recently attracted much attention because of their pronounced quasi-one-dimensional nature (an exchange between the adjacent spins along the hexagonal axis is much stronger than that between the neighbors in the plane). Experiments[6–8] have demonstrated large deviations of the staggered magnetization and susceptibility in such compounds from the results of classical calculations, and thus the importance of quantum fluctuations. Therefore, they are also simple examples of quantum helimagnets, which have also been recently studied in the literature.[9–11] Chubukov[12] and Tanaka *et al.*[13] calculated the classical spin reversal process in a magnetic field (see also Ref. 14) and the antiferromagnetic resonance (AFMR) spectra in the hexagonal antiferromagnets of $CsNiCl_3$ type on the basis of the six-sublattice model. Even for six sublattices it is very difficult to handle the problem in this way, and essentially no explicit analytical result can be obtained for the spin-wave spectrum. Our motivation in the present work was to develop another, simpler, and more general procedure for finding the classical ground-state spin configuration, which would allow one to perform reasonably unsophisticated spin-wave calculations. We employ the procedure which is similar to that used by Nagamiya[3] and Kaplan.[4] Actually, it is an extension of the iterative method first suggested by Cooper *et al.*[15] for easy-plane, almost ferromagnetic spirals, which is valid for a general helimagnet. However, keeping in mind its application to the commensurate antiferromagnets, we will not discuss here such effects as changes in the ordering wave vector **Q** caused by the anisotropy and magnetic field that occur for a general **Q** position. These problems, as well as the incommensurate-commensurate transitions, were treated in the framework of the phenomenological approach.[16,17]

We start from the following Hamiltonian which describes the low-temperature magnetic properties of a large class of crystals in which the spins of the magnetic ions are localized at the lattice points

$$\hat{\mathcal{H}}=\sum_{i,j} J_{ij}\mathbf{S}_i\mathbf{S}_j+D\sum_i (S_i^z)^2-\gamma\mathbf{H}\sum_i S_i. \qquad (1)$$

Here the first term is the Heisenberg exchange interaction, the second term describes the lowest-order uniaxial anisotropy, and the last term is the Zeeman term (γ is the gyromagnetic ratio). The anisotropy arises from the magnetic spin–spin and spin–orbit interactions. Hence, the constant D incorporates a small relativistic ratio $(v/c)^2$, where v is the average velocity of the electron in the atom, and c is the velocity of light. Therefore, this ratio is usually considered to be small in comparison with the exchange coupling constants J_{ij} which are of the electrostatic origin. This condition, however, can be violated in the quasi-low-dimensional materials, where the coupling along some directions is indirect and weak and the corresponding constants J_{ij} are very small. Such cases, as well as the singlet ground state systems in which D is intrinsically very large, are interesting in themselves, but in the present paper we will always imply the condition $|D|\ll|J_{ij}|\neq 0$. We also restrict our consideration to the case in which the summation in (1) is performed over a single Bravais lattice with total N sites.

In the spin-wave theory the ground state and the excitation spectrum of the quantum Hamiltonian (1) are calculated in the quasiclassical approximation based on the $1/S$ expansion. The starting point of such a calculation is to find an equilibrium spin configuration which minimizes the ground-state energy $E_{\text{g.s.}}$ in the classical limit $S\rightarrow\infty$. A rather complete spin-wave theory describing its low-temperature properties has been developed for the case of the isotropic Heisenberg Hamiltonian.[9–11] However, the problem becomes extremely complicated if the anisotropy existing in real compounds is included together with the magnetic field. Even for the classical helimagnets very little general results were therefore actually obtained. Some particular cases were successfully treated in Refs. 3 and 15, but they lead to rather involved expressions (partially because an attempt to account for the sixfold anisotropy, in addition to the easy-plane anisotropy, made the problem much more complicated). Here we shall present the calculations of the ground state spin configuration of the Hamiltonian (1) for different orientations of the magnetic field, taking into account the anisotropy (but not the field) as a first-order perturbation of the exchange interaction.

2. FAILURE OF THE WEAK CONDITION

It is known that the classical ground state for a system of equivalent spins on a simple Bravais lattice in the exchange approximation is a magnetic spiral described by wave vector $\mathbf{Q}$ (including ferromagnetism ($\mathbf{Q}=0$) and antiferromagnetism ($\mathbf{Q}=\mathbf{K}/2$) as particular cases). The rigorous proof of this result can be found by solving a mathematical problem for the absolute minimum of the function (1) which depends on $3N$ classical variables S_i^α under N conditions

$$S_i^2=S^2,\quad \forall i \tag{2}$$

imposed on the lengths of the classical spins. This problem is solved by introducing N Lagrange multipliers λ_i. Switching to the Fourier representation because of the lattice translational symmetry, we obtain the following system of equations for spin configuration which minimizes the Hamiltonian

$$NJ_{\mathbf{k}}\mathbf{S}_{\mathbf{k}}+\mathbf{e}_z DS_{\mathbf{k}}^z-\sum_{\mathbf{k}'}\lambda_{\mathbf{k}'}\mathbf{S}_{\mathbf{k}-\mathbf{k}'}=\frac{1}{2}\gamma\mathbf{H}\delta_{\mathbf{k},0},$$

$$\sum_{\mathbf{k}'}\mathbf{S}_{\mathbf{k}'}\mathbf{S}_{\mathbf{k}-\mathbf{k}'}=S^2\delta_{\mathbf{k},0}. \tag{3}$$

Here $J_{\mathbf{k}}$, $\mathbf{S}_{\mathbf{k}}$, and $\lambda_{\mathbf{k}}$ are the (lattice) Fourier transforms of the functions J_{ij}, $\mathbf{S}_i$, and λ_i, respectively; $\mathbf{e}_z$ is a unit vector along z direction, and $\delta_{\mathbf{k},0}$ is a three-dimensional Kronekker symbol. From (3) we easily obtain the classical ground-state energy

$$\frac{1}{N}E_{\text{g.s.}}=\lambda_0-\frac{\gamma}{2}\mathbf{H}\mathbf{S}_0. \tag{4}$$

There are two points to note here. First, (3) is a complicated inhomogeneous system of nonlinear equations, for which no general solution is found. Secondly, the resultant ground-state energy explicitly depends only on the values λ_0 and $\mathbf{S}_0$ (on the latter only if $\mathbf{H}\neq 0$), so all $\lambda_{\mathbf{k}}$ and $\mathbf{k}\neq 0$ seem to be irrelevant. This fact encourages one to look for the solution for which the Lagrange multipliers take the form $\lambda_{\mathbf{k}}=\lambda_0\delta_{\mathbf{k},0}$ and the system is largely linearized. Evidently, such procedure is equivalent to taking into account only one of the N conditions given by the second relation in (3) or replacing (2) by the so-called "weak" condition $\Sigma_i\mathbf{S}_i^2 = NS^2$. This is a standard approach, which was generally formalized by Lyons and Kaplan.[5] Then the solution is easily found to be a single $\mathbf{Q}$ helix

$$\mathbf{S}_{\mathbf{k}}=\mathbf{S}_0\delta_{\mathbf{k},0}+\mathbf{S}_{\mathbf{Q}}\delta_{\mathbf{k},\mathbf{Q}}+\mathbf{S}_{\mathbf{Q}}^*\delta_{\mathbf{k},-\mathbf{Q}},$$

$$\lambda_0=NJ_{\mathbf{Q}}=N\min\{J_{\mathbf{k}}\}, \tag{5}$$

which is actually a correct result for $D\geqslant 0$ and $\mathbf{H}$ along the z axis. One can easily choose vectors $\mathbf{S}_0$ and $\mathbf{S}_{\mathbf{Q}}$ in (5), so that all N "strong" conditions (2) are satisfied for any D and for the direction of the field. Thus, according to the Luttinger–Tisza theorem, the resultant spin configuration is the solution of (3), i.e., it minimizes the energy (1) under the "strong" conditions (2).

Unfortunately, except for the above important case, simple solutions obtained in this way appear to be physically meaningless. For example, for any negative (easy-axis) anisotropy the magnetic structure at $H=0$ is predicted (with the help of the "weak" condition) to be a collinear configuration parallel to the z axis with spins of varying length.[4] This, in particular, does not describe the experimental data in $CsNiCl_3$ and related compounds in which the easy-axis anisotropy is known to fix the spin plane parallel to z and to distort slightly the perfect exchange 120° structure which is initially described by a helix with $\mathbf{Q}=(1/3,1/3,1)$ in reciprocal lattice units. [In fact, the "incorrect" collinear phase also appears in $CsNiCl_3$ in the narrow temperature interval between two split T_N (Ref. 18)].

The reason for this contradiction and inconsistency of the trick with the "weak" condition is the following. It is easy to see that by weakening the conditions on the spin length we obtain some extra minima that do not satisfy (2).

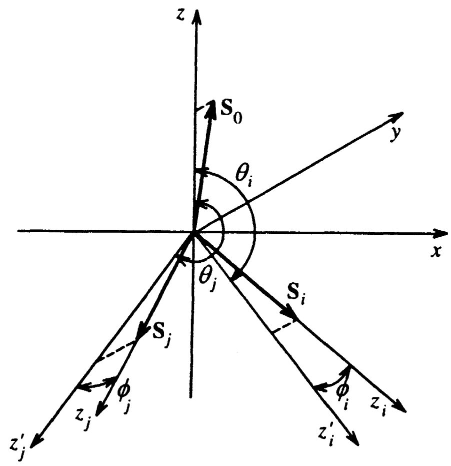

FIG. 1. Spin configuration of the distorted exchange spiral and transformation to the local axes.

These artifacts are thrown away by verifying all the conditions explicitly. However, some extrema of the initial problem (3) are *lost* in this way. This can be visualized by considering the energy surface (1) in the space of spin components cut by the "physical" manifold (2). If the "real" solution lies at one of the intersection points, it will be inevitably missed by employing the "weak" condition, since it is not an extremum of the function (1) in the extended space.

3. TRANSFORMATION TO THE LOCAL AXES

Having found the correct classical ground-state spin configuration, we proceed with the spin-wave theory defining the local coordinate axes $x_i y_i z_i$, so that (classically) the spin at each site points in the z_i direction in the ground state. The spin operators are then transformed to these axes and decompose into a series of Bose operators using the Holstein-Primakoff transformations. Since the local z_i axis is a classical equilibrium direction of each spin, linear in spin deviations (i.e., in operators $\hat{a}_i^+, \hat{a}_i$), terms are absent in the resultant decomposition of the Hamiltonian. Starting from this point, we can develop a slightly different procedure to find the classical ground state of the Hamiltonian (1) and its decomposition into a series of Bose operators. First, we transform the spins from the crystallographic xyz frame to the local coordinate axes $x_i y_i z_i$. In general, this is done by two rotations: first, by some angle θ_i around the y axis, and then by some angle ϕ_i around the new x'_i axis, as shown in Fig. 1. In the resultant local frame the spin operators are

$$\tilde{\mathbf{S}}_i = \|\mathbf{T}_{2,i}\| \cdot \|\mathbf{T}_{1,i}\| \cdot \mathbf{S}_i = \|\mathbf{T}_i\| \cdot \mathbf{S}_i, \tag{6}$$

where the transformation matrices are, as usual, defined by

$$\|\mathbf{T}_{1,i}\| = \begin{Vmatrix} \cos\theta_i & 0 & -\sin\theta_i \\ 0 & 1 & 0 \\ \sin\theta_i & 0 & \cos\theta_i \end{Vmatrix},$$

$$\|\mathbf{T}_{2,i}\| = \begin{Vmatrix} 1 & 0 & 0 \\ 0 & \cos\phi_i & -\sin\phi_i \\ 0 & \sin\phi_i & \cos\phi_i \end{Vmatrix}. \tag{7}$$

Transformed Hamiltonian then takes the form

$$\mathscr{H} = \sum_{i,j} J_{ij} \tilde{\mathbf{S}}_i \|\mathbf{T}_{ij}^{(\mathrm{ex})}\| \tilde{\mathbf{S}}_j + D \sum_i \tilde{\mathbf{S}}_i \|\mathbf{T}_i^{(a)}\| \tilde{\mathbf{S}}_i - \gamma \mathbf{H} \sum_i \|\mathbf{T}_i\|^{-1} \tilde{\mathbf{S}}_i. \tag{8}$$

Here the matrices $\|\mathbf{T}_i^{(a)}\|$ and $\|\mathbf{T}_{ij}^{(\mathrm{ex})}\|$ which define the bilinear, in spin components, forms coming from anisotropy and exchange, respectively, are given by

$$\|\mathbf{T}_{ij}^{(a)}\| = \begin{Vmatrix} \sin^2\theta_i & \sin\phi_i \sin\theta_i \cos\theta_i & -\cos\phi_i \sin\theta_i \cos\theta_i \\ \sin\phi_i \sin\theta_i \cos\theta_i & \sin^2\phi_i \cos^2\theta_i & -\sin\phi_i \cos\phi_i \cos^2\theta_i \\ -\cos\phi_i \sin\theta_i \cos\theta_i & -\sin\phi_i \cos\phi_i \cos^2\theta_i & \cos^2\phi_i \cos^2\theta_i \end{Vmatrix}, \tag{9}$$

$$\|\mathbf{T}_{ij}^{(\mathrm{ex})}\| = \begin{Vmatrix} \cos\theta_{ij} & \sin\theta_{ij}\sin\phi_j & -\sin\theta_{ij}\cos\phi_j \\ -\sin\theta_{ij}\sin\phi_i & \cos\phi_i\cos\phi_j + \cos\theta_{ij}\sin\phi_i\sin\phi_j & \cos\phi_i\sin\phi_j - \cos\theta_{ij}\sin\phi_i\cos\phi_j \\ \sin\theta_{ij}\cos\phi_i & \sin\phi_i\cos\phi_j - \cos\theta_{ij}\cos\phi_i\sin\phi_j & \sin\phi_i\sin\phi_j + \cos\theta_{ij}\cos\phi_i\cos\phi_j \end{Vmatrix}, \tag{10}$$

where $\theta_{ij} = \theta_i - \theta_j$. The angles θ_i and ϕ_i must be chosen in such a way that one can exclude terms $\sim \tilde{S}_i^x \tilde{S}_j^z$ and $\sim \tilde{S}_i^y \tilde{S}_j^z$ which give rise to the linear, in $\hat{a}_i^+$, $\hat{a}_i$, contribution to the Hamiltonian (8). This leads to the following system of $2N$ equations):

$$\sum_i 2J_{ij} \cos\phi_i \sin\theta_{ij} = D\cos\phi_j \sin 2\theta_j + \tilde{\gamma} H_x \cos\theta_j - \tilde{\gamma} H_z \sin\theta_j,$$

$$\sum_i 2J_{ij} (\cos\phi_j \sin\phi_i - \sin\phi_j \cos\phi_i \cos\theta_{ij}) = D \sin 2\phi_j \cos^2\theta_j - \tilde{\gamma} H_x \sin\phi_j \sin\theta_j + \tilde{\gamma} H_y \cos\phi_j - \tilde{\gamma} H_z \sin\phi_j \cos\theta_j, \tag{11}$$

where $\tilde{\gamma} = \gamma/S$. Evidently, for arbitrary D and H this system for the angles θ_i and ϕ_i cannot be resolved. As was mentioned above, only the cases $D = 0$ and $D > 0$, $\mathbf{H} \| \mathbf{z}$ can be treated explicitly.[1,11] However, we can begin with the

Heisenberg exchange Hamiltonian in a magnetic field as a zero approximation and then look for the corrections to it, expanding them to a given power of D. A very similar approach has been developed by Andreev and Marchenko[19] in the framework of the phenomenological Lagrangian theory. They showed rather simple and general solution of the problem in the leading order of perturbation.

First, we consider a more complicated case of the easy-axis anisotropy which causes the most general distortion of the exchange structure.

4. EASY-AXIS ANISOTROPY ($D<0$)

Without loss of generality, we can choose the magnetic field to lie in the yz plane. Furthermore, in this case the "correct" unperturbed helix at $H=0$ should contain the z axis. We therefore look for the solution of (11) in the form

$$\left\{\begin{matrix} \theta_i=\mathbf{Qr}_i+\delta\theta_i \\ \phi_i=\phi_0+\delta\phi_i \end{matrix}\right\}, \quad \sin\phi_0=\frac{\tilde{\gamma}H}{2(J_0-J_{\mathbf{Q}})}. \tag{12}$$

In most cases we shall consider only the leading (i.e., the lowest order in D) corrections $\delta\theta_i$, $\delta\phi_i$. Thus, substituting (12) into (11), it is sufficient to expand the trigonometric functions to the first order in $\delta\theta_i$, $\delta\phi_i$. The following equations for the deviations

$$\begin{aligned}
&\sum_i 2J_{ij}(\cos\phi_0\cos(\mathbf{Qr}_{ij})\delta\theta_i-\sin\phi_0\sin(\mathbf{Qr}_{ij})\delta\phi_i)\\
&\quad=2J_{\mathbf{Q}}\cos\phi_0\delta\theta_j+D\cos\phi_0\sin(2\mathbf{Qr}_j)\\
&\quad\quad-\tilde{\gamma}H_z[\cos(\mathbf{Qr}_j)\delta\theta_j+\sin\mathbf{Qr}_j)]\\
&\sum_i 2J_{ij}[(\cos^2\phi_0+\sin^2\phi_0\cos(\mathbf{Qr}_{ij}))\delta\phi_i\\
&\quad+\sin\phi_0\cos\phi_0\sin(\mathbf{Qr}_{ij})\delta\theta_i]\\
&\quad=2(J_0\sin^2\phi_0+J_{\mathbf{Q}}\cos^2\phi_0)\delta\phi_j+D\sin\phi_0\cos\phi_0\\
&\quad\quad\times[1+\cos(2\mathbf{Qr}_j)]\\
&\quad\quad-[\tilde{\gamma}H_y\sin\phi_0+\tilde{\gamma}H_z\cos\phi_0\cos(\mathbf{Qr}_j)]\delta\phi_j\\
&\quad\quad+\tilde{\gamma}H_z\sin\phi_0\sin(\mathbf{Qr}_j)\delta\theta_j
\end{aligned} \tag{13}$$

are rather easy to solve. Being convoluted with $J_{ij}\cos(\mathbf{Qr}_{ij})$, the variations $\delta\theta_i$ and $\delta\phi_i$ should give the trigonometric functions $\cos(\mathbf{Qr}_j)$, $\sin(\mathbf{Qr}_j)$, $\cos(2\mathbf{Qr}_j)$, $\sin(2\mathbf{Qr}_j)$, etc., which would cancel the right-hand side of equation (11). Thus, they should be sought in the form of a decomposition into a sum

$$\delta\theta_i=\Sigma_n[\alpha_n\cos(n\mathbf{Qr}_i)+\beta_n\sin(n\mathbf{Qr}_i)],$$

where the order of the coefficients α_n and β_n is H^n and $|D|^{n/2}$.

Here we should make one very important remark. Instead of (12), one can look for the solution of (11) in the form $\theta_i=\psi+\mathbf{Qr}_i+\delta\theta_i$. It differs from the first one in a rotation of the arbitrarily chosen "first" spin, and thus of the whole spin structure by an angle ψ within the helix plane. Evidently, in our approximation $\sim D$ all such structures will have the same energy, as it was the case for the pure exchange. This fact reflects the remaining continuous degeneracy of the ground state with respect to the described rotations. Moreover, by performing corresponding expansions it can be shown in a straightforward algebraic way that such degeneracy exists in all orders in H, $\sqrt{|D|}$ unless the condition $n\mathbf{Q}\equiv 0$ is satisfied for some integer n. Such a condition means that the spin structure is a commensurate spiral and can be described in terms of n sublattices. In this case the continuous degeneracy of the spin rotation within the helix plane is lifted in the order H^n and $|D|^{n/2}$. The ground state preserves only n-fold degeneracy corresponding to the arbitrary choice of the "first" spin, i.e., to the rotations of all spins in the initial exchange structure by an angle $(2\pi m)/n$, within the helix plane. In the case of the incommensurate spiral, where this additional discrete degeneracy is absent, the continuous degeneracy of the ground state is not destroyed.

This fact can be proved by very general arguments. Evidently, perturbation of the n-sublattice exchange spiral by adding $D\Sigma_i\cos^2(\psi+\theta_i)$ or $H\Sigma_i\cos(\psi+\theta_i)$ to the exchange Hamiltonian is invariant with respect to the transformation $\psi\rightarrow\psi+(2\pi m)/n$. Thus, the ground-state energy should not contain harmonics below $\cos n\psi$ and $\sin n\psi$. Hence, its Taylor expansion with respect to the perturbation parameters D and H depends explicitly on the angle ψ in the terms of $n/2$ and n order, respectively. Another way to explain this fact is based on the exchange symmetry arguments.[19] Exchange structure with n sublattices has an n-fold rotation axis perpendicular to the spin plane. Thus, the lowest order perturbation to the exchange energy, which has appropriate symmetry and which lifts such rotations should be proportional to $\cos n\psi$. In the case of the uniaxial anisotropy, which already contains the twofold symmetry, it is achieved in the $n/2$ order of perturbation, while for the magnetic field the nth order is needed. With the continuous degeneracy destroyed, the corresponding Goldstone mode gains frequency induced by an "effective field," $|D|^{n/2}$ or H^n; it is of the order $|D|^{n/4}$ and $H^{n/2}$, respectively.

This situation was closely studied in the hexagonal $CsNiCl_3$-type antiferromagnets, which are our reference in the present paper. It was shown[20,21] that in the easy-axis case the structure with $\psi=0$, given by (12), is stabilized by the anisotropy in the third order, and the magnon gap $\sim|D|^{3/2}$ appears. For the easy-plane antiferromagnetic helix in the transverse field the situation is different (as we will discuss later) and the structure with $\psi=\pi/2$ is chosen.[12,14] Hereafter, we shall consider the solution (12), which is stabilized by the easy-axis anisotropy $D<0$, with two principal orientations of the magnetic field.

4.1. Magnetic field perpendicular to the z axis

This orientation gives rise to the most general distortion of the exchange helix. Deviations of the angles satisfying (13) are found in the form

$$\delta\theta_i=\vartheta\sin(2\mathbf{Qr}_i), \quad \vartheta=\frac{D}{J_{3\mathbf{Q}}-J_{\mathbf{Q}}}+\frac{D}{J_{2\mathbf{Q}}-J_{\mathbf{Q}}}\tan^2\phi_0,$$

$$\delta\phi_i=\frac{D}{2(J_0-J_{\mathbf{Q}})}\tan\phi_0+\varphi\cos(2\mathbf{Qr}_i),$$

$$\varphi=\frac{D}{J_{2\mathbf{Q}}-J_{\mathbf{Q}}}\tan\phi_0. \tag{14}$$

Note that according to these expressions, an anisotropy causes a small modulation of the helix turn angle $\mathbf{Qr}_i$ that propagates as $2\mathbf{Qr}_i$. This naturally results in the appearance of the components at the wave vectors $\pm 2\mathbf{Q}$ and $\pm 3\mathbf{Q}$ in the Fourier transform of the ground-state spin arrangement, in agreement with the earlier results of Nagamiya *et al.*[3] and Cooper and Elliott.[15] Another point to be mentioned here is that small variations (14) that were treated in the linear approximation contain terms which increase with the magnetic field (the most drastic is the second term in ϑ proportional to H^2). So despite being valid up to very high fields these expressions fail near the spin-flip transition at

$$\tilde{\gamma}H\sim 2(J_0-J_{\mathbf{Q}})\left(1+\frac{D}{2(J_{2\mathbf{Q}}-J_{\mathbf{Q}})}\right).$$

In fact, this failure indicates that there is a phase transition to a new spin arrangement which is somewhat similar to the "fan" structure that was considered in detail by Nagamiya *et al.*[3] A simple way to study this transition and the behavior of the system in the vicinity of the spin flip in the framework of the present approach is to expand the angles ϕ_i in small deviations from $\pi/2$. This method was applied in Ref. 14, where such transitions were studied in the triangular antiferromagnet $CsMnBr_3$.

The classical ground-state energy corresponding to the found spin arrangement can be easily calculated from (7)–(10):

$$E_{\mathrm{g.s.}}=NS^2(J_{\mathbf{Q}}+\tfrac{1}{2}D-(J_0-J_{\mathbf{Q}}+\tfrac{1}{2}D)\sin^2\phi_0)$$
$$=NS^2(J_{\mathbf{Q}}+\tfrac{1}{2}D)-\tfrac{1}{2}N\chi_\perp(\gamma H)^2, \tag{15}$$

where we introduce the transverse spin susceptibility per site as follows:

$$\chi_\perp=\frac{1}{2(J_0-J_{\mathbf{Q}})-D}$$
$$=\frac{1}{2(J_0-J_{\mathbf{Q}})}\left(1+\frac{D}{2(J_0-J_{\mathbf{Q}})}\right)+O(D^2). \tag{16}$$

Spin structures are usually verified in neutron scattering experiments by measuring the intensities of the magnetic Bragg peaks. At each $\mathbf{k}$, they are proportional to the square of the absolute value of the corresponding Fourier transform of the spin density $\mathbf{S}_{\mathbf{k}}$, which therefore can be called a Bragg amplitude. Thus, for comparison with experiment it is important to calculate nonzero Bragg amplitudes $\mathbf{S}_{\mathbf{k}}$ that result from solution (14). Introducing the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_y$ along the x and y directions, we obtain

$$\mathbf{S}_0=\mathbf{e}_yS\sin\phi_0\left(1+\frac{D}{2(J_0-J_{\mathbf{Q}})}\right)=\chi_\perp(\gamma\mathbf{H}),$$

$$\mathbf{S}_{\mathbf{Q}}=\frac{1}{2}(\mathbf{e}_z-i\mathbf{e}_x)S\cos\phi_0\left(1-\frac{D}{2(J_0-J_{\mathbf{Q}})}\tan^2\phi_0\right)$$
$$-(\mathbf{e}_z+i\mathbf{e}_x)S\cos\phi_0\left(\frac{D}{4(J_{3\mathbf{Q}}-J_{\mathbf{Q}})}+\frac{D}{2(J_{2\mathbf{Q}}-J_{\mathbf{Q}})}\tan^2\phi_0\right),$$

$$\mathbf{S}_{2\mathbf{Q}}=\mathbf{e}_yS\sin\phi_0\frac{D}{2(J_{2\mathbf{Q}}-J_{\mathbf{Q}})},$$

$$\mathbf{S}_{3\mathbf{Q}}=(\mathbf{e}_z-i\mathbf{e}_x)S\cos\phi_0\frac{D}{4(J_{3\mathbf{Q}}-J_{\mathbf{Q}})}. \tag{17}$$

Calculating the Bragg intensities $I_{\mathbf{k}}\sim|\mathbf{S}_{\mathbf{k}}|^2$, we should keep in mind that the vectors $\mathbf{S}_{\mathbf{Q}}$ and $\mathbf{S}_{3\mathbf{Q}}$ have a nonzero imaginary part.

4.2. Magnetic field along the *z* axis; low fields

On the one hand, this case seems to be less complicated, since the distortion of the exchange helix (5), which occurs only at low fields $H<H_c\sim\sqrt{|D|J}$, leaves the spins coplanar. This means that $\phi_i=0$, and that the second equation in (11) and (13) is automatically satisfied. The first one is reduced to a rather simple relation, from which the small variations $\delta\theta_i$ to the leading order in D and H are

$$\delta\theta_i=\vartheta\sin(2\mathbf{Qr}_i)+\eta\sin(\mathbf{Qr}_i),\quad \vartheta=\frac{D}{J_{3\mathbf{Q}}-J_{\mathbf{Q}}},$$

$$\eta=-\frac{\tilde{\gamma}H}{J_0+J_{2\mathbf{Q}}-2J_{\mathbf{Q}}}. \tag{18}$$

It is evident from these expressions that leading order is insufficient in our approximation. To be consistent in the first order in the anisotropy constant D, it is necessary to perform the expansions to the second power of $\eta\sim H\leqslant\sqrt{|D|J}$. Such improved procedure leads to the following corrected expression for ϑ:

$$\vartheta=\frac{D}{J_{3\mathbf{Q}}-J_{\mathbf{Q}}}+\eta^2\left(\frac{J_{2\mathbf{Q}}-J_{\mathbf{Q}}}{J_{3\mathbf{Q}}-J_{\mathbf{Q}}}-\frac{1}{4}\right). \tag{19}$$

The corresponding ground-state energy and parallel susceptibility per spin are

$$E_{\mathrm{g.s.}}=NS^2\left(J_{\mathbf{Q}}+\frac{1}{2}D-\frac{(\tilde{\gamma}H)^2}{4(J_0+J_{2\mathbf{Q}}-2J_{\mathbf{Q}})}\right),$$

$$\chi_\parallel=\frac{1}{2(J_0+J_{2\mathbf{Q}}-2J_{\mathbf{Q}})}. \tag{20}$$

We can rewrite $\eta=-2\chi_\parallel(\tilde{\gamma}H)$. The Bragg amplitudes can also be easily calculated:

$$\mathbf{S}_0=\mathbf{e}_zS\frac{\tilde{\gamma}H}{2(J_0+J_{2\mathbf{Q}}-2J_{\mathbf{Q}})}=\chi_\parallel\gamma\mathbf{H},$$

$$\mathbf{S}_{\mathbf{Q}}=\frac{1}{2}(\mathbf{e}_z-i\mathbf{e}_x)S\left(1-\frac{D+\eta^2(J_{2\mathbf{Q}}-J_{\mathbf{Q}})}{2(J_{3\mathbf{Q}}-J_{\mathbf{Q}})}\right)$$
$$+i\mathbf{e}_xS\left[\frac{1}{2}\eta^2-\frac{D+\eta^2(J_{2\mathbf{Q}}-J_{\mathbf{Q}})}{J_{3\mathbf{Q}}-J_{\mathbf{Q}}}\right],$$

$$S_{2\mathbf{Q}} = -(\mathbf{e}_z - i\mathbf{e}_x) S \frac{\tilde{\gamma} H}{4(J_0 + J_{2\mathbf{Q}} - 2J_\mathbf{Q})}$$

$$= -\frac{1}{2} \chi_\| \gamma H (\mathbf{e}_z - i\mathbf{e}_x),$$

$$S_{3\mathbf{Q}} = (\mathbf{e}_z - i\mathbf{e}_x) S \frac{D + \eta^2 (J_{2\mathbf{Q}} - J_\mathbf{Q})}{4(J_{3\mathbf{Q}} - J_\mathbf{Q})}. \tag{21}$$

Actually, we can calculate the homogeneous component of the spin density S_0 (i.e., the magnetization) with better accuracy than in (21), i.e., to $\sim |D|^{3/2}$. To be completely consistent in this order, we should add to $\delta\theta_i$ terms like $\beta \sin(3\mathbf{Q}\mathbf{r}_i)$ with $\beta \sim |D|^{3/2}$. However, kept in the first order, these terms will not contribute to $\mathbf{S}_0$. For our purpose, it is therefore sufficient to take $\delta\theta_i$ in the form (18) and retain more terms in the subsequent expansions. In this way we obtain the following correction to $\mathbf{S}_0$:

$$\mathbf{S}_0 = \chi_\| \gamma \mathbf{H} \left[1 - \chi_\| D \left(3 - 4 \frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{J_{3\mathbf{Q}} - J_\mathbf{Q}} \right) + (4\tilde{\gamma} H) 2 \chi_\|^3 (J_{2\mathbf{Q}} - J_\mathbf{Q}) \left(\frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{J_{3\mathbf{Q}} - J_\mathbf{Q}} - \frac{1}{2} \right) \right]. \tag{22}$$

This expression describes the leading nonlinearity of the magnetization and can be important for comparison with the experiment.

4.3. Magnetic field along *z* axis; high fields

This case is the simplest and the most well-known case. At some critical field $H_c \sim \sqrt{|D| J}$, the usual spin-flip transition takes place and the structure becomes the same as that of the magnetized exchange helix (5):

$$\mathbf{S}_\mathbf{k} = \mathbf{e}_z S \sin\phi_0 \delta_{\mathbf{k},0} + (\mathbf{e}_x + i\mathbf{e}_y) S \cos\phi_0 \delta_{\mathbf{k},\mathbf{Q}} + (\mathbf{e}_x - i\mathbf{e}_y) S \cos\phi_0 \delta_{\mathbf{k},-\mathbf{Q}}. \tag{23}$$

Here

$$E_{\text{g.s.}} = N S^2 \left(J_\mathbf{Q} - \frac{(\tilde{\gamma} H)^2}{4(J_0 - J_\mathbf{Q} + D)} \right),$$

$$\sin\phi_0 = \frac{\tilde{\gamma} H}{2(J_0 - J_\mathbf{Q} + D)}. \tag{24}$$

This is correct up to the complete saturation, where the corresponding spin-flip field is defined by the condition $\sin\phi_0 = 1$. Comparing expressions (20) and (24) for the ground-state energy, we obtain the following universal formula for the spin-flip field H_c:

$$\tilde{\gamma} H_c = \sqrt{2|D|(J_0 - J_\mathbf{Q}) \left(1 + \frac{J_0 - J_\mathbf{Q}}{J_{2\mathbf{Q}} - J_\mathbf{Q}} \right)}. \tag{25}$$

5. EASY-PLANE ANISOTROPY ($D>0$)

Actually the spin reversal in the magnetic field for the easy-plane Heisenberg helimagnet demonstrates no new features, aside from those described in the previous section. All spin configurations occurring in this case have their exact analogs in the easy-axis helimagnet. It is sufficient to establish a correspondence between the cases.

The simplest case is one in which the magnetic field is directed along the z axis. It is exactly the same as that in Sec. 4.3 and is described by the same expressions [(23) and (24)]. Here we must keep in mind that the constant D is now positive.

If the field is applied in the easy plane, e.g., along the x axis, in low fields the spin arrangement is analogous to that described in Sec. 4.2. This correspondence, however, is not as direct as the one above. Since all spins lie in the plane, the solution of system (11) is obtained with $\theta_i = \pi/2$ and $\phi_i = \mathbf{Q}\mathbf{r}_i + \delta\phi_i$. The first equation is then automatically satisfied, while the second one for ϕ_i becomes exactly the same as the equation for θ_i in the case $D<0$, $\mathbf{H}\|\mathbf{z}$, $H<H_c$, but with $D=0$. Since now the symmetry breaking in the (easy) spin plane is different, we will consider two possible solutions.

A. First, we can assume $\delta\phi_i = \eta \sin(\mathbf{Q}\mathbf{r}_i) + \vartheta \sin(2\mathbf{Q}\mathbf{r}_i)$, which is exactly the same as that considered in Sec. 4.2. As a result, we find η and ϑ given by (18) and (19) (of course, for $D=0$). It is evident from transformation (7) that the resultant spin Fourier components are given by the same expressions, (21), where $\mathbf{e}_x$ and $\mathbf{e}_z$ are replaced by $\mathbf{e}_y$ and $\mathbf{e}_x$, respectively. Such solution can be stabilized by the magnetic field in the case of a nearly ferromagnetic helix, where the spins tend to align along the field direction.

B. Consider the solution in the form $\delta\phi_i = \pi/2 + \eta \cos(\mathbf{Q}\mathbf{r}_i) - \vartheta \sin(2\mathbf{Q}\mathbf{r}_i)$. The parameters η and ϑ are again given by (18) and (19). As was discussed above, in our approximation this solution is degenerate with the previous one and differs from it only in the phase multipliers in the Bragg amplitudes:

$$\mathbf{S}_0 = \chi_\| \gamma \mathbf{H} \left[1 + (4\tilde{\gamma} H)^2 \chi_\|^3 (J_{2\mathbf{Q}} - J_\mathbf{Q}) \left(\frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{J_{3\mathbf{Q}} - J_\mathbf{Q}} - \frac{1}{2} \right) \right],$$

$$\mathbf{S}_\mathbf{Q} = \frac{1}{2} (\mathbf{e}_y + i\mathbf{e}_x) S \left(1 - \eta^2 \frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{2(J_{3\mathbf{Q}} - J_\mathbf{Q})} \right) - \frac{1}{2} e_y S \eta^2 \left[\frac{1}{2} - \frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{J_{3\mathbf{Q}} - J_\mathbf{Q}} \right],$$

$$\mathbf{S}_{2\mathbf{Q}} = (\mathbf{e}_x - i\mathbf{e}_y) S \frac{\tilde{\gamma} H}{4(J_0 + J_{2\mathbf{Q}} - 2J_\mathbf{Q})} = \frac{1}{2} \chi_\| \gamma H (\mathbf{e}_x - i\mathbf{e}_y),$$

$$\mathbf{S}_{3\mathbf{Q}} = -(\mathbf{e}_y + i\mathbf{e}_x) S \eta^2 \frac{J_{2\mathbf{Q}} - J_\mathbf{Q}}{4(J_{3\mathbf{Q}} - J_\mathbf{Q})}. \tag{26}$$

As was shown by Chubukov[12] for the case of a six-sublattice commensurate helimagnet $CsMnBr_3$, it is a type-**B** structure which is stabilized by the magnetic field in the sixth order. The corresponding frequency $\sim H^3$ appears in the magnetic resonance spectrum. The magnetic field makes favorable this solution for an antiferromagnetic spiral, since in this case it tends to establish the spins perpendicular to its direction. In the general case of the n-sublattice antiferromagnetic spiral, this structure (26) is stabilized by the magnetic field in the nth order, as discussed in Sec. 4.

Above the spin-flip transition, which occurs at $\mathbf{H}\perp\mathbf{z}$ and $D>0$ at the same field H_c given by (25), the spin structure becomes exactly the same as that discussed in Sec. 4.1 (assuming $\mathbf{H}\|\mathbf{y}$) and is described by the same expressions (for $D>0$).

6. CONCLUSION

Here we presented the first complete treatment of the classical spin reversal process for a weakly uniaxial microscopic Heisenberg Hamiltonian (1), consistent up to the first order in the anisotropy constant D. Previously, each particular case of this Hamiltonian was considered in the framework of the sublattice model. Many examples of such approach are found in the calculations of spin reversals in the hexagonal "triangular" antiferromagnets by Chubukov,[12] Tanaka,[13] and Abarzhi *et al.*[14] In addition to the loss of generality, in nontrivial cases such calculations are rather difficult and result in very complicated formulas, depending on the particular form of the exchange interactions. Thus, in most cases the spectrum of the spin waves and magnetic resonance cannot be calculated in explicit analytical expressions on the basis of the sublattice model. In contrast, the results obtained in the present paper are quite general and rather simple. They provide a solid basis for the subsequent spin-wave calculations and estimates of the fluctuational contributions to the ground-state energy that can be very important (even for the choice of the correct ground state). Although the spin structures were calculated only for two principal orientations of the magnetic field, we can easily generalize for any orientation. For this purpose, we must retain the relevant terms in (11), (13) and mix the $\sin(\mathbf{Qr}_i)$ and $\cos(\mathbf{Qr}_i)$ rotations in the angle variations $\delta\phi_i$, $\delta\theta_i$. Thus, for example, some results reported in Ref. 14 can be obtained in a more general way.

As was already mentioned, starting with the exchange approximation, one can treat the magnetic structures and the long-wavelength spin dynamics in the framework of the phenomenological Lagrangian theory proposed by Andreev and Marchenko.[19] For the systems described by Hamiltonian (1) its leading relativistic corrections are equivalent to our treatment of the anisotropy. This theory has the advantage of being less subjected to the corrections due to fluctuation effects (the mean-field ground state is chosen better). However, it also has two substantial disadvantages in comparison with the microscopic theory developed here. First, it is not aimed at calculation of the spin density distribution and the excitations spectrum in the whole Brillouin zone. Thus, it cannot describe neutron scattering experiments, which are the most sensitive tool for studying spin systems. Secondly, to reproduce the higher-order effects like the nonlinearity of the magnetization given by (22) such theory requires introduction of the additional phenomenological constants. Of course, the microscopic theory worked out in the classical approximation presented here is far from being perfect and requires a serious treatment of quantum corrections. This point, as well as the calculations of the magnon spectra, will be the subject of our further studies.

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