

# Nonlinear dynamics of highly anisotropic spin-1 magnetic materials

V. S. Ostrovskii

*Institute of Physics, Academy of Sciences of the Ukrainian SSR*

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The completeness of the dynamical description of magnetic substances with spins  $S > 1/2$  is discussed. It is shown that the minimum number of dynamical variables (and, consequently, of equations for them) necessary to consider all the interactions allowed by the magnitude of the spin adequately is equal to  $4S$ . A set of four equations that furnish a self-consistent description of the dynamics of an  $S = 1$  magnetic material is explicitly derived on the basis of the single-site coherent states for the  $SU(3)$  Lie group. Physical situations are considered whose most important feature is not the orientational motion of the magnetization vector, but the dynamics of the quadrupole degrees of freedom, which constitute an important element of the total dynamics. Solutions are specifically obtained for: 1) a linearized quadrupole wave describing the oscillations of the magnetization modulus and the quadrupole-tensor axes, 2) a domain wall whose magnetization modulus varies continuously in the absence of spin rotation as it moves in an external field, and 3) a magnetic vortex with a two-dimensional order parameter that vanishes on the axis. The solutions found are essentially quantum-mechanical and do not arise in the semiclassical approach. In the static limit the set of equations obtained describes a self-consistent ground state, and also solves the problem of the diagonalization of an arbitrary single-site Hamiltonian for  $S = 1$ .

## 1. INTRODUCTION

The solution of the Landau-Lifshitz equation (LLE)<sup>1</sup> has in the last few years allowed significant progress to be made in the understanding of the nonlinear dynamics of magnetically ordered crystals (see, for example, the reviews in Refs. 2 and 3). This equation furnishes a self-consistent description of the orientational dynamics of a magnetization vector  $\mathbf{s}(r)$  satisfying the condition  $|\mathbf{s}(\mathbf{r})| = \text{const}$ , and is, strictly speaking, valid for spin systems with<sup>4</sup>  $S = 1/2$ . The LLE yields reasonable results for arbitrary  $S$  values as well when the relativistic interactions are weak compared to the exchange interactions,<sup>5</sup> and we can approximate them by effective anisotropy fields. However, even in this limiting case a more exact description often becomes necessary. With that end in view nonmodel theories of linear magnetization-vector dynamics that use only symmetry arguments were developed.<sup>6–8</sup>

At the same time a number of examples are known in which the fields and interactions of higher multipole order, which stem from incompletely quenched orbital motion, are comparable to, or even much stronger than, the magnetic field and the bilinear exchange. The static and resonance properties of such crystals are quite pronounced, and in many respects both differ qualitatively from the analogous properties of slightly anisotropic magnetic materials and depend essentially on the specific magnitude of the spins of the magnetic ions. In certain cases there are especially striking differences between the properties of systems with integral<sup>9–11</sup> and half-integral<sup>11,12</sup> spins, and the acoustic (magnon) branches actually interact with the optical branches of comparable energy, the number of the latter being also dependent on  $S$ . These and many other characteristics<sup>13</sup> are a consequence of the competition among the contribution to

the ground state and the elementary excitation spectrum from those interactions of different natures which cannot be reduced just to effective fields, i.e., to functions of only the magnetization. Equally justified allowance for all the competing interactions, irrespective of their natures, can be made naturally on the basis of quantum theory. In this case we can, with the aid of a small number of parameters of an appropriately chosen model, quite satisfactorily explain a large amount of experimental data on such highly anisotropic magnetic materials as for instance  $\text{CoF}_2$  (Ref. 12). Thus far this theory appears mostly worked out,<sup>9–17</sup> but the methods that have been developed for it are not suited for the description of highly excited inhomogeneous states, i.e., for the typical problems of nonlinear dynamics.

For the investigation of nonlinear problems, it is desirable to have for the dynamics a description similar to the LLE, i.e., equations of motion that are as physically clear (which makes it much easier to work with them) and as well justified from the standpoint of microtheory as the LLE for  $S = 1/2$ . This implies first and foremost that their completeness, i.e., that they be able to incorporate any interactions allowed by the magnitude of the spins, as well as the self-consistency of the interspin interaction when exact allowance is made for the single-site interactions, which is in accord with the idea underlying the quantum approach.<sup>1)</sup> A key question here is the question of the number of dynamical variables necessary for a complete description. The answer to it can be obtained by expanding an arbitrary spin state in terms of some complete basis, e.g.,

$$\Psi(t) = \sum_{M=-S}^{-S} a_M(t) |S, M\rangle. \quad (1)$$

We can, taking account of the normalization condition and

the arbitrariness in the choice of the general phase in (1), see from this that the evolution of the state is governed by the  $4S$  independent real parameters entering into the coefficients  $a_M$ . It is natural to choose as more convenient dynamical variables  $4S$  independent quantities from the set of  $4S(S+1)$  mean—over the state (1)—operators constituting the  $SU(2S+1)$  Lie algebra (e.g., the irreducible tensors  $O_{k,q}$ ,  $k=1, \dots, 2S$ ). These simple arguments indicate, in particular, the fundamental character of the difficulties that arise in the phenomenological description of highly anisotropic magnetic substances on the basis of functions of the three magnetization-vector components: even for  $S=1$  the minimum number of dynamical variables is equal to four. Let us emphasize that here (and below) the topic under discussion is “the minimally complete” description of the dynamics, i.e., a description with the aid of pure states, the region of applicability of which is precisely the region of greatest interest to us: it is precisely at  $T \ll T_c$  that the quantum effects caused by the freezing out of additional (nondipole) degrees of freedom most dramatically manifest themselves. The elucidation of the properties of these degrees of freedom and their effect on the dynamics of the spin system is the main aim of the present paper.<sup>2)</sup>

In the present paper we investigate the  $S=1$  case, which is the simplest case for which we can go beyond the LLE. In Sec. 2 we explicitly derive and discuss a set of four equations that completely describe the dynamics of an arbitrary magnetic material with  $S=1$ . In Sec. 3 we consider examples of nondipole dynamics: a quadrupole wave and a domain boundary in a biaxial ferromagnet, as well as the magnetic vortex with a variable modulus  $s(\mathbf{r})$ . The results are discussed in Sec. 4.

## 2. SPIN DYNAMICS EQUATIONS FOR $S=1$

Keeping in mind the self-consistent description of the dynamics of a spin system, we first consider the description of the state of an isolated spin at the site  $n$ . In the  $S=1$  case let us choose as the complete set of operators of the  $SU(3)$  Lie algebra the three components of the spin  $S_n^i$  and the five independent components of the symmetric tensor

$$Q_n^{ij} = \frac{1}{2}(S_n^i S_n^j + S_n^j S_n^i) - \delta_{ij} S(S+1)/3.$$

We shall seek the four independent dynamical variables and the equations for them by going over into each site's proper moving coordinate system (MCS):

$$\Psi = R\Psi', \quad O = RO'R^{-1}, \quad R = \Pi R_n, \quad R_n = \exp(-i\Phi_n S_n), \quad (2)$$

which we define by the following conditions for the averages  $\langle \dots \rangle_0 \equiv \langle \psi'_n | \dots | \psi'_n \rangle$ :

$$\langle S_n^i \rangle_0 = 0, \quad \langle S_n^\alpha \rangle_0 = 0, \quad \langle Q_n^{\xi\xi} \rangle_0 = 0 \quad (3)$$

(here and below the Greek indices pertain only to the MCS:  $\alpha, \beta = \xi, \eta, \zeta$ , and their subscript  $n$  will be dropped:  $\alpha \equiv \alpha_n$ ,  $\xi \equiv \xi_n$ , etc.).

The first two conditions in (3) specify the form of the function with which the averaging is carried out up to an arbitrary phase:

$$\psi'_n = \cos g_n |1\rangle + \sin g_n |-1\rangle, \quad (4)$$

while the third ensures the reality of its coefficients. There then remains only one free parameter, namely, the parameter  $g_n$ , which can be directly related to the average spin length  $s_n = \langle S_n^\xi \rangle_0$  and the quadrupole moment  $q_n = \langle Q_n^{\xi\xi} - Q_n^{\eta\eta} \rangle$ :

$$s_n = \cos 2g_n, \quad q_n = \sin 2g_n, \quad s_n^2 + q_n^2 = 1; \quad (5)$$

for the remaining average quantities we have the identities

$$\langle Q_n^{\xi\xi} \rangle_0 = \langle Q_n^{\eta\eta} \rangle_0 = 0, \quad \langle Q_n^{\xi\xi} \rangle = 1/3. \quad (6)$$

Thus, it is natural to choose as the four independent variables describing the state of the spin at the site  $n$  one of the quantities  $(g_n, s_n, q_n)$  and the three parameters of the unitary transformation  $R_n$  that effect the transition into the proper MCS. If we represent the transformation  $R_n$  by three Euler rotations (which is convenient in specific calculations), then two angles  $(\varphi_n, \vartheta_n)$  specify, as usual, the orientation of the spin  $\langle s_n \rangle_0$ , while the third angle  $(\gamma_n)$  describes the rotation of the quadrupole moment about this vector. This rotation, which is not a real rotation in the semiclassical description, is, as will be seen below, an important element of the dynamics of an anisotropic magnetic material.

We shall derive the dynamical equations for these variables from the equations of motion (in the MCS) for the operators  $O'_n = S_n^\alpha, Q_n^{\alpha\beta}$  averaged over the state (4):

$$i\hbar \frac{d}{dt} \langle O'_n \rangle_0 = \langle [O'_n, \tilde{\mathcal{H}}'_n + i\hbar R_n^{-1} R_n] \rangle_0, \quad (7)$$

where

$$\tilde{\mathcal{H}}'_n = \mathcal{H}'_n + \sum_m \langle \psi'_m | \mathcal{H}'_{nm} | \psi'_m \rangle, \quad (8)$$

and  $\mathcal{H}'_n, \mathcal{H}'_{nm}$  are respectively the single-site- and pair-interaction operators in the MCS. Using the relations (3)–(6), we arrive, after some calculations,<sup>21</sup> at the required system of equations:

$$\dot{s}_n - 2q_n \bar{A}_n^{\xi\eta} = 0, \quad (9a)$$

$$\Omega_n^{\xi} + \tilde{\mathcal{H}}_n^{\xi} - \frac{s_n}{2q_n} (\bar{A}_n^{\xi\xi} - \bar{A}_n^{\eta\eta}) = 0, \quad (9b)$$

$$\Omega_n^{\eta} + \tilde{\mathcal{H}}_n^{\eta} + \frac{s_n}{1-q_n} \bar{A}_n^{\eta\xi} = 0, \quad (9c)$$

$$\Omega_n^{\xi} + \tilde{\mathcal{H}}_n^{\xi} + \frac{s_n}{1+q_n} \bar{A}_n^{\xi\xi} = 0. \quad (9d)$$

[The remaining four equations can be obtained from (9a)–(9d) and (3)–(6).] Here the  $\Omega_n^\alpha = \dot{\Phi}_n^\alpha$  are the components of the angular velocity in the MCS:

$$d\Phi_n^{\xi} = d\vartheta_n \sin \gamma_n - d\varphi_n \sin \vartheta_n \cos \gamma_n, \quad (10)$$

$$d\Phi_n^{\eta} = d\vartheta_n \cos \gamma_n + d\varphi_n \sin \vartheta_n \sin \gamma_n,$$

$$d\Phi_n^{\xi} = d\gamma_n + d\varphi_n \cos \vartheta_n;$$

while

$$\tilde{\mathcal{H}}_n^\alpha = H_n^\alpha + \sum_m J_{nm}^{\alpha\xi} s_m, \quad \bar{A}_n^{\alpha\beta} = A_n^{\alpha\beta} + \sum_{m,\nu} K_{nm}^{\alpha\beta\nu\nu} \langle Q_m^{\nu\nu} \rangle_0 \quad (11)$$

are the effective magnetic and quadrupole fields, which can

be expressed in terms of the constants of the Hamiltonian of the general (for  $S = 1$ ) form:

$$\mathcal{H} = - \sum_n \{H_n^i S_n^i + A_n^{ij} Q_n^{ij}\} - \frac{1}{2} \sum_{nm} \{J_{nm}^{ij} S_n^i S_m^j + K_{nm}^{ijkl} Q_n^{ij} Q_m^{kl}\}, \quad (12)$$

and the parameters of the transformation  $R_n$ :

$$\begin{aligned} H_n^\alpha &= \sum_i H_n^i u_n^{i\alpha}, & J_{nm}^{\alpha\beta} &= \sum_{ij} J_{nm}^{ij} u_n^{i\alpha} u_m^{j\beta}, \\ A_n^{\alpha\beta} &= \sum_{ij} A_n^{ij} u_n^{i\alpha} u_n^{j\beta}, \\ K_{nm}^{\alpha\beta\gamma\mu} &= \sum_{ijkl} K_{nm}^{ijkl} u_n^{i\alpha} u_n^{j\beta} u_m^{k\gamma} u_m^{l\mu}, \end{aligned} \quad (13)$$

where  $u_n^{i\alpha}(\varphi, \vartheta, \gamma)$  is the well-known transformation matrix for the vector

$$S_n^i = \sum_\alpha u_n^{i\alpha} S_n^\alpha.$$

Let us discuss the physical meaning and some characteristics of the above system of equations. It is shown in the Appendix that the equations (9a)–(9d) provide for the spin motion a natural adiabatic description that differs from the usual adiabatic approximation<sup>22</sup> in that, under the appropriate initial condition  $\Psi_n(t_0) = T_n(t_0)\psi_n''$ , the other eigenfunctions of the operator,  $\mathcal{H}_n^{\text{eff}}$  do not contribute to the evolution of  $\psi_n(t)$ . For typical problems of the dynamics of magnetically ordered crystals the fulfillment of this condition is guaranteed by the fact that the equations (9a)–(9d) which determine the parameters of the transformation  $T_n(t)$ , describe in the static limit a self-consistent equilibrium state that can be considered to be the initial condition at  $t_0 \rightarrow -\infty$  for both the nonlocalized and localized solutions. It can be directly verified that the state  $\psi_n = T_n|1\rangle_n = R_n\psi_n'$  possesses all the properties of a generalized coherent state,<sup>23</sup> corresponding in our case to the dynamical  $SU(3)$  symmetry group. This state corresponds to a point in the four-dimensional factor space  $SU(3)/H$ , where the stationary subgroup  $H$  of the state  $\psi_n' = |1\rangle$  consists of unitary rotations with generators  $Q_n^{zz}, S_n^z, S_n^x - 2Q_n^{xz}, S_n^y - 2Q_n^{yz}$ . It is also easy to verify that, in the MCS, the function (4) minimizes the uncertainty relations for the sets of three operators  $(S_n^\xi, S_n^\eta, S_n^\zeta)$ ,  $(Q_n^{\xi\eta}, Q_n^{\xi\xi} - Q_n^{\eta\eta}, S_n^\zeta)$  etc. The relation between the  $T_n$  and  $R_n$  transformations of the function  $\psi_n, \psi_n'$ , and  $\psi_n''$  is given in the Appendix. Naturally,  $T_n$  can be parametrized in any other way, but the selection of space rotations gives the equations transparent physical meaning, as was done above in the derivation of (9a)–(9d).

[We note that the transformation  $T_n$  does not completely diagonalize  $\mathcal{H}_n^{\text{eff}}$ , but distinguishes only one eigenvector, the evolution of which is described by Eqs. (9a)–(9d). But the solution of this system of four equations reduces the problem of the final diagonalization to the reduction of a  $2 \times 2$  matrix, which is easily carried out by means of two successive rotations about the vector  $\psi_n''$  with the generators  $S_n^x - 2Q_n^{xz}$  and  $S_n^y - 2Q_n^{yz}$ . Note that in the case of the static problem the equilibrium state is determined by a smaller number of parameters (see Ref. 10)<sup>3</sup>.]

We can get some idea about the deviation of Eqs. (9a)–(9d) from the LLE and about the nature of the dynamics of  $S = 1$  spins by analyzing the terms due to the bilinear exchange. Going over in the difference equations to the continuum description, limiting ourselves, as usual, to expansion terms of up to the second derivatives in the coordinates, and introducing the fields  $(s_n, \varphi_n, \vartheta_n, \gamma_n) \rightarrow (s, \varphi, \vartheta, \gamma) \equiv [s(\mathbf{r}), \dots]$ , we obtain

$$\begin{aligned} \sum_\delta J_{n,n+\delta}^{\alpha\zeta} s_{n+\delta} &\rightarrow \{\delta_{\alpha\zeta} s J_0 + s j_0^{\alpha\zeta}\} \\ &+ \left\{ s \sum_\delta J_\delta \sum_\nu \varepsilon_{\zeta\nu} (\delta\nabla) \Phi^\nu + \delta_{\alpha\zeta} \sum_\delta J_\delta (\delta\nabla) s \right\} \\ &+ \left\{ \frac{1}{2} s \sum_\delta J_\delta \sum_i u^{i\alpha} (\delta\nabla)^2 u^{i\zeta} + \sum_\delta J_\delta (\delta\nabla) s \sum_\nu \varepsilon_{\zeta\nu} (\delta\nabla) \Phi^\nu \right. \\ &\left. + \frac{1}{2} \delta_{\alpha\zeta} \sum_\delta J_\delta (\delta\nabla)^2 s \right\}, \end{aligned} \quad (14)$$

where

$$J_\delta^{ij} = J_{n,n+\delta}^{ij} = J_\delta + j_\delta^{ij}, \quad J_0^\delta = \sum_\delta J_\delta, \quad j_0^{\alpha\zeta} = \sum_{\delta ij} j_\delta^{ij} u_0^{i\alpha} u_0^{j\zeta}.$$

If there is a center of inversion, the expression in the second brackets vanishes, and the one in the third brackets can, when the crystal contains two axes or symmetry planes, be reduced to the simpler form

$$J' a^2 \left\{ \frac{1}{2} s \sum_i u^{i\alpha} \Delta u^{i\zeta} + \sum_\nu \varepsilon_{\zeta\nu} (\nabla \Phi^\nu) (\nabla s) + \frac{1}{2} \delta_{\alpha\zeta} \Delta s \right\}, \quad (15)$$

where  $J'$  is the so-called inhomogeneous-exchange constant:  $2J_\delta \ll J' \ll J_0$ .

The meaning of each term in (14) and (15) is clear from its structure. The fact that the majority of these terms enter only into one or two pairs of equations, namely, (9a), (9b) or (9c), (9d), is especially noteworthy. In particular, the principal (isotropic) part  $sJ_0$  of the homogeneous exchange occurs only in (9b), and this applies also to the last terms in the second and third brackets in (14), whereas the penultimate terms in them are contained only in (9c) and (9d). (The anisotropy field  $s j_0^{\alpha\zeta}$  and the purely orientational part of the inhomogeneous exchange are identical to the corresponding terms in the LLE.) These differences are some of the consequences of the fundamentally different natures of the two types of motion that are involved in magnetic dynamics, and are, in the general case, coupled to each other. In certain situations, however, the degrees of freedom corresponding to them turn out to be independent, or else only one of them actually exists. Thus, in the limit of weak quadrupole interactions, i.e., for  $\bar{A}_n^{\alpha\beta} \rightarrow 0$ , we find from (9a) and (9b) that  $q_n \rightarrow 0, s_n \rightarrow 1$ , and  $\dot{s}_n \rightarrow 0$ ; the angle  $\gamma_n$  can then be chosen so that the pair (9c) and (9d) go over into the LLE, describing only the orientational dynamics—the important dynamics in this case—of a magnetization vector with constant modulus. Of greatest interest to us is the other case, in which for one reason or another it is just the orientational spin motion that does not occur or is of secondary importance, and the dominant motion is the one described by Eqs.

(9a) and (9b). The remaining part of the paper is devoted to the analysis of these equations and the motion corresponding to them.

### 3. THE QUADRUPOLE SPIN DYNAMICS OF A FERROMAGNET

In the present section we shall consider certain situations in which the dominant role is played by the pair of equations (9a) and (9b), i.e., situations that are, in a sense, opposite to those that can at least qualitatively be described with the aid of the LLE. We shall consider two such possibilities in the particular case of a biaxial ferromagnet with a single-ion anisotropy (SA):

$$\mathcal{H} = - \sum_n \{ HS_n^z + A Q_n^{zz} + B(Q_n^{xx} - Q_n^{yy}) \} - \frac{1}{2} \sum_{nm} J_{nm} S_n S_m, \quad (16)$$

in the ground state of which (when  $A > B$  and  $H = 0$ ) the spins have length  $s_n = s_0 = (1 - b^2)^{1/2}$ , where  $b = B/J_0$ ,<sup>9</sup> and are oriented along  $Z$ . As follows from the explicit form of Eqs. (9a)–(9d), in the absence of spin motion connected with deviations of the orientation of the spins from this axis (or for small amplitudes of such deviations) the motion described by the first pair,

$$\hbar \dot{s}_n + 2Bq_n \sin 2\gamma_n = 0, \quad (17a)$$

$$\hbar \dot{\gamma}_n - B \frac{s_n}{q_n} \cos 2\gamma_n + \sum_{\delta} J_{\delta} s_{n+\delta} + H = 0, \quad (17b)$$

can be considered to be independent. The physical meaning of these equations is quite clear. In them the magnetization  $s_n$  acts like an essentially quadrupole dynamical variable; its variation is determined by the angle  $\gamma_n$  of rotation of the quadrupole moment about the  $Z$  axis, and the rate of this rotation is in turn given by the deviation of the magnetization [or the quantity  $q_n = (1 - s_n^2)^{1/2}$ ] from the equilibrium state.

#### Quadrupole wave

The simplest example of such a motion is the linearized quadrupole wave, which, as will be shown below, corresponds to the exchange branch in the elementary excitation spectrum of a ferromagnet with  $S = 1$ , just as a spin wave corresponds to the acoustic (magnon) branch. By differentiating (17a) with respect to the time we go over to the second-order equation

$$\hbar \ddot{s}_n + 4B^2 s_n + 4Bq_n \left( \sum_{\delta} J_{\delta} s_{n+\delta} + H \right) \cos 2\gamma_n = 0, \quad (18)$$

where  $\cos 2\gamma_n$  is defined in (17a). We shall seek the solution in the form

$$s_n(t) = \bar{s} + \sigma \cos(\mathbf{k}\mathbf{n} - \omega t), \quad (19)$$

where  $\bar{s}$  is the homogeneous steady-state solution to the system (17a) and (17b):  $\dot{s}_n = \dot{\gamma}_n = 0$ ,  $s_n = \bar{s}$ . Linearizing (18) with respect to  $s_n - \bar{s}$  under the condition that  $\sigma \ll \bar{q}^2 = 1 - \bar{s}^2$ , we find the dispersion relation

$$\hbar \omega(\mathbf{k}) = 2J_0 [ (b/\bar{q})^2 - b\bar{q}\Gamma(\mathbf{k}) ]^{1/2}, \quad (20)$$

$$\Gamma(\mathbf{k}) = z^{-1} \sum_{\delta} \cos(\mathbf{k}\delta), \quad z = \sum_{\delta} 1.$$

For the angle  $\gamma_n$  we have

$$\gamma_n = (\hbar \omega \sigma / 2J_0 b \bar{q}) \sin(\mathbf{k}\mathbf{n} - \omega t). \quad (19')$$

In weak fields  $h = H/J_0$

$$\bar{s} = s_0 + \chi_{\parallel} h, \quad \chi_{\parallel} = b^2 / (1 - b^2) \quad (21)$$

and

$$\hbar \omega(\mathbf{k}) = 2J_0 [ (1 + \kappa h)^2 - b^2 (1 - \kappa h) \Gamma(\mathbf{k}) ]^{1/2}, \quad (22)$$

where  $\kappa = 1/s_0 = (1 + \chi_{\parallel})^{1/2}$ .

This dispersion relation can be obtained on the basis of the microscopic theory (see Ref. 9) by expanding (16) in terms of the basis operators  $|\varphi_i\rangle_n \langle \varphi_k|$ :

$$\mathcal{H}_2 = \sum_n [ 2(1 + h/\bar{s}) J_0 ] B_n^+ B_n - \frac{1}{2} \sum_{n\delta} J_{\delta} \bar{q}^2 (B_n^+ B_m + B_n B_m + \text{H.c.}). \quad (23)$$

Here we have written out only the operators,  $B_n = |\varphi_0\rangle_n \langle \varphi_2|$  and  $B_n^+ = |\varphi_2\rangle_n \langle \varphi_0|$ , that are essential to excitations of the type in question, and are responsible for the transitions between the states  $|\varphi_0\rangle_n \cos \bar{g} |1\rangle + \sin \bar{g} | -1\rangle$ ,  $|\varphi_2\rangle = -\sin \bar{g} |1\rangle + \cos \bar{g} | -1\rangle$ , where  $\cos 2\bar{g} = \bar{s}$ . The Hamiltonian (23) describes the optical (exchange) branch of the excitation spectrum of the ferromagnet, a branch which is also called the longitudinal branch, and is associated with the collective tunneling of the spins between the states  $|1\rangle$  and  $| -1\rangle$ , which does not require the surmounting of an orientational barrier of height  $\sim A$ . This branch interacts only with a longitudinal variable field, and has, when  $A > J_0$ , an activation energy smaller than the gap in the spectrum of the acoustic magnons, so that it can be excited independently of the latter excitations both resonantly and thermally. When the dispersion relations in the two approaches coincide, the nature of the excitations is more clearly revealed in the description of the dynamics with the aid of coherent states, and, what is most important, in the present approach no difficulties arise in the investigation of essentially nonlinear problems.

#### The domain wall

Another situation in which only the first pair of equations, i.e., Eqs. (9a) and (9b), need to be considered arises in the analysis of the domain wall (DW) structure in the model (16) with  $A - B > J_0$ . Indeed, the width  $\Delta = a [J_0 / (A - B)]^{1/2}$  of a Bloch- or Néel-type DW decreases with increasing anisotropy (in the process of spins inside the wall "shorten"<sup>24</sup>), and the rotation of the magnetization becomes energetically disadvantageous at  $A - B > J_0$  (i.e., when  $\Delta < a$ ), so that Eqs. (9c) and (9d) can again be ignored. In order to investigate the structure and dynamics of the DW in this case, let us go over in (18) to the continuum description, and introduce the dimensionless variables

$\bar{x} = 2^{1/2}x/a$  and  $\tau = 2(B/J_0)^{1/2}t/\hbar$ , as well as the relaxation term  $\alpha'b^{-1/2}s_\tau \cos 2\gamma$  [in Eq. (17b) to it will correspond the term  $\alpha'J_0 \sin 2\gamma_n$ ]:

$$s_{\tau\tau} + bs - \cos 2\gamma \{gs_{\bar{x}\bar{x}} + q(s+h) - \alpha'b^{-1/2}s_\tau\} = 0. \quad (24)$$

An analysis shows,<sup>25</sup> the free ( $H=0, \alpha'=0$ ) DW motion corresponding to the boundary conditions  $s(\mp\infty) = \pm s_0$  is possible at velocities  $u < u_k = b^{1/2} [u = V\hbar(2BJ_0a^2)^{-1/2}$  is the dimensionless velocity]. Furthermore, the DW width is equal to  $\Delta_0 = 2a(1-b)^{-1/2}$  at  $u=0$ , and tends to the finite limit  $\Delta_k = a2^{1/2}$  as  $u \rightarrow u_k$ , whence we obtain the condition of applicability of the continuum approximation:  $1-b \ll 1, u \ll u_k$ . We have up to and including terms of second order in  $w^2 = u^2/u_k^2$  the solution

$$\frac{\xi - \xi_0}{(1-w^2)^{1/2}} = \frac{b}{s_0} \text{Arsh} \left\{ \frac{ss_0}{(1-s_0^2)^{1/2} - b} \right\} + \arcsin s, \quad (25)$$

$$\sin 2\gamma = \frac{u}{u_k} \frac{ds/d\xi}{(1-s^2)^{1/2}} \approx \frac{u}{u_k} \frac{s_0^2}{1+b \text{ch}(s_0\xi)}, \quad (26)$$

which describes a moving DW ( $\xi = \bar{x} - u\tau$ ) having a magnetization (25) that varies in magnitude, and accompanying its localized quadrupole-moment rotation wave (26), the amplitude of which vanishes at  $u=0$  (dynamical soliton). By taking account of the fact that  $b \sim q \lesssim 1$ , we can give a slightly less exact solution for  $s$ :

$$s = s_0 \text{sh} \xi / (\text{ch} \xi + b), \quad \xi = s_0(\xi - \xi_0)/(1-w^2)^{1/2}.$$

Notice that the expression (25) with  $w$  replaced by  $u$  is the exact solution to the simplified equation

$$s_{\bar{x}\bar{x}} - s_{\tau\tau} + s - bs/(1-s^2)^{1/2} = 0. \quad (27)$$

The rest energy of such a DW,  $E_0 = 2^{-1/2}J_0(\arcsin s_0 - bs_0) \approx 2^{1/2}J_0s_0^3/3$  for  $s_0 \ll 1$ , is much smaller than the DW energy in the Ising model with  $s_n = \text{const}$ .

Let us consider the DW motion in an external field. Neglecting, for simplicity sake, the longitudinal susceptibility (21), we find the acceleration of a particle of mass<sup>25</sup>  $m_0 = (2^{1/2}\hbar^2J_0s_0^3/6aB)$ , having the DW coordinate  $X_0 = Vt$ :

$$\frac{dV}{dt} = -\frac{1}{m_0a} \frac{\partial}{\partial X_0} \int \{-Hs(x-X_0)\} dx = \frac{6 \cdot 2^{1/2}B^2aH}{\hbar^2J_0s_0^2} \quad (28)$$

(in dimensionless quantities  $du/d\tau = 3bh/s_0^2$ ).

In the presence of dissipation, i.e., for  $\alpha' \neq 0$ , the motion stabilizes at some velocity  $V_{\alpha'}(H)$ , which we can determine from the condition  $dE/d\tau = 0$ . Computing the energy of the crystal, and using Eqs. (17a) and (17b), we have

$$dE/d\tau \sim \left\{ h \int s_\tau dx - \alpha'b^{-1/2} \int (s_\tau^2/q) dx \right\} = 0,$$

whence, taking account of the fact that  $\partial/\partial\tau = -V\hbar(4BJ_0)^{-1/2}\partial/\partial x$ , we find

$$V_{\alpha'}(H) = \frac{3 \cdot 2^{1/2}aBH}{J_0\hbar s_0^2 \alpha'} \left( \text{or } u_{\alpha'}(h) = \frac{3b^{1/2}}{s_0^2} \frac{h}{\alpha'} \right). \quad (29)$$

Allowance for the discreteness of the lattice leads to the appearance of an additional periodic potential with barrier height  $\Delta E = J_0[b - 3/2(2b^2)^{2/3}]$ .

## Magnetic vortex

Let us consider one more example of the situation in which the variability of the modulus of  $s_n$  essentially changes the nature of the solution; specifically, let us discuss the structure of a vortex in an easy-plane ferromagnetic with a single-ion anisotropy:

$$\mathcal{H} = \sum_n DQ_n^{zz} - \frac{1}{2} \sum_{nm} J_{nm}(S_n^x S_m^x + S_n^y S_m^y + p S_n^z S_m^z). \quad (30)$$

As is well known,<sup>2,26</sup> in the model with  $|s| = \text{const}$  the vortex-energy minimum is attained when the magnetization at the vortex core leaves the plane. This departure, which has been substantiated for weakly anisotropic crystals, occurs in the case of strong single-ion anisotropy only when  $S$  is half-integral. Let us, without dwelling at length on the analysis, note that in the present case the magnetization does not, on account of the Kramers theorem, vanish anywhere, and that in the limit when  $D \gg J_0$  all the changes in the modulus of  $s_n$  can be taken into account in the simple model with  $S_{ef} = 1/2$ , an anisotropic  $g$  factor ( $g_\perp/g_\parallel = S + 1/2$ ), and an anisotropic exchange [ $J_\perp/J_\parallel = (S + 1/2)^2/p$ ].

The situation is entirely different in the case of integral  $S$ . Let us illustrate this for  $S=1$ . As we saw in the preceding example, the magnitude  $s_n$  of the magnetization is the result of the competition between the effective field acting on the spin and the single-ion tensor component perpendicular to this field, and should vanish wherever the field vanishes. This conclusion is valid for any models with  $S=1$ , and in the appropriate generalizations for any integral  $S$ . In particular, for the model (30) the nontrivial solution  $s_n = s_\infty \neq 0$  to the homogeneous problem is energetically advantageous only when  $d = D/(2J_0) < 1$ :  $s_\infty = (1-d^2)^{1/2}$  (Ref. 10); for  $d > 1$  the ground state is a singlet. At the same time, for spins oriented along the  $Z$  axis the relation  $s_n = 1$  holds for any  $d$  (irrespective of what causes the orientation). Returning to the problem of the vortex, we can say that the state in which the magnetization remains in the  $XY$  plane and decreases as the distance to the vortex center decreases [i.e., the state in which  $\vartheta = \pi/2$  and  $s(\mathbf{r}) \rightarrow 0$  as  $\mathbf{r} \rightarrow 0$ ] is admissible in the  $S=1$  case,<sup>4)</sup> leaves the gradient energy finite, and, more importantly, is energetically more advantageous than the state with  $\vartheta(0) = 0$  or  $\pi$  and  $s(0) = 1$  in some region of the parameters (e.g., when  $d \lesssim 1$  and  $s_\infty \ll 1$ ); in this case the transition from one type of vortex to another when the parameter  $d$  is varied should clearly be a first-order transition.

Let us find this solution. Going over in (9a)–(9d) to the continuum description, and setting the polar angle  $\vartheta = \pi/2$ , we obtain from (9c) and (9d) the equations

$$\begin{aligned} \hbar\dot{\varphi} \sin \gamma + J_0a^2 \cos \gamma [^{1/2}S\Delta\varphi + (\nabla s)(\nabla\varphi)] &= 0, \\ -\hbar\dot{\varphi} \cos \gamma + J_0a^2 \sin \gamma [^{1/2}S\Delta\varphi + (\nabla s)(\nabla\varphi)] &= 0, \end{aligned} \quad (31)$$

whence  $\dot{\varphi} = 0$ , i.e., the vortex is “frozen-in.” This conclusion does not change when allowance is made for an external field in the  $XY$  plane, so that the vortex motion should still be accompanied by the orientation of the spins along the  $Z$  axis.

For the static problem we obtain from (9a) and (9b) the equation

$$ds/q + s^{-1/2} a^2 s (\nabla\varphi)^2 + 1/2 a^2 \Delta s = 0. \quad (32)$$

Let us seek the solution for a vortex in the form  $(\nabla s) \cdot (\nabla\varphi) = 0$ ,  $s = s(r)$ . Then  $\varphi = \varphi(\lambda)$ , and from (31) we have  $\varphi = \nu\lambda + \varphi_0$ , where  $r$  and  $\lambda$  are the cylindrical coordinates of a point in the plane. Next, going over to the variables  $\rho = s_\infty r/a$  and  $\sigma = s/s_\infty$ , we obtain for  $\sigma(\rho)$  the equation:

$$\sigma_{\rho\rho} + \frac{1}{\rho} \sigma_\rho + \left[ \frac{2}{s_\infty^2} \left( 1 - \frac{d}{q} \right) - \frac{\nu^2}{\rho^2} \right] \sigma = 0. \quad (33)$$

Being interested in the  $s_\infty \ll 1$  case, we shall consider the asymptotic solutions in the vicinity of the vortex axis and at the periphery.

As  $\rho \rightarrow 0$ , (33) goes over into the Bessel equation, and

$$\sigma_<(\rho) = J_\nu(\rho) \approx \frac{1}{\nu!} \left( \frac{\rho}{2} \right)^\nu = \left( \frac{r}{r_\nu} \right)^\nu, \quad (34a)$$

where  $r_\nu = 2a(\nu!)^{1/\nu} s_\infty^{-1}$  is the characteristic scale: the vortex-core radius, which, for  $s_\infty \ll 1$ , can be much greater than the interatomic distance  $a$ .

For  $\rho \rightarrow \infty$  the solution has the form

$$\sigma_>(\rho) \approx 1 - \nu^2/2\rho^2. \quad (34b)$$

[In the case of the ordinary vortex, the orientational inhomogeneity decreases more rapidly:  $\vartheta = \pi/2 - c'e^{-r}/r^{1/2}$  (Refs. 2 and 26).] The vortex energy is equal to

$$E_c = J_0 \frac{\pi s_\infty^2}{2d^2} \left\{ \nu^2 \ln \left| \frac{\rho_R}{\rho'} \right| + \frac{1}{4} d^2 \rho'^2 \right\}, \quad (35)$$

where  $\rho_R = R s_\infty/a$ ,  $R$  is the vortex radius, and  $\rho'$  is a quantity of the order of the vortex-core radius, as which it is reasonable to choose the distance to the point where the functions  $\sigma_<$  and  $\sigma_>$  are matched (for  $\nu = 1$ ,  $\rho' = 2^{1/2}$ , and for  $\nu = 2$ ,  $\rho' = 2$ ).

Let us, in conclusion of this section, emphasize that the solutions obtained above—the quadrupole wave, the DW, and the vortex—are due to the possibility of the contraction of the spin in the pure state (i.e., at  $T = 0$ ), and are thus essentially quantum effects, of which those (the DW, the vortex) in which  $s(r)$  vanishes are admissible only in the case of integral  $S$ . This, of course, does not exclude the possibility that similar objects exist as a result of the purely statistical shrinkage of the mean moment, a process which usually occurs<sup>28</sup> as  $T \rightarrow T_c$  (a special case is the  $X$ - $Y$  model, for which the vanishing of the moment on the vortex axis also occurs purely statistically at  $T \rightarrow 0$ ). In all such cases the magnitude of the spins does not play any role.

#### 4. CONCLUSION

The need to consider the possibility of the variation of the magnetization modulus has been repeatedly pointed out in the literature. As has been shown above, the magnetization modulus does not in itself constitute a new dynamical degree of freedom, and is even not a pure vector variable: its sites of the quadrupole fields and interactions. Without allowance for the latter in a theory that operates only with functions of the magnetization vector, the magnitude of the elasticity can act only as a relaxation variable.<sup>29</sup> In the gen-

eral case all the degrees of freedom, the number of which is determined by the magnitude of the spins, participate in the dynamics; we can eliminate some of them from the analysis by replacing the multipole interactions with effective anisotropy fields only under the assumption that these interactions are weak. The system of equations (9a)–(9d) obtained here allowed us to take account of all the necessary dynamical degrees of freedom of a magnetic material for the simplest ( $S = 1$ ) case that permits us to go beyond the purely orientational dynamics described by the LLE. The examples discussed above, in which the dominant role is played by the pair of equations (9a) and (9b), besides being of interest in their own right, allow us to get some idea about that aspect of the dynamics of magnetically ordered crystals with spins  $S > 1/2$  which usually remains in the shadow, although it exerts a significant influence on all the observable properties of anisotropic spin systems. The method of self-consistent coherent states, which is used in the present paper for the  $S = 1$  case, allows us to obtain a complete description of the dynamics and statics for any  $S$ , adequately (and equitably) taking account of all the admissible interactions. Here there is no need to express the spin operators in terms of Bose or other convenient operators, and there does not arise the problem of taking account of the so-called kinematic interaction—a pathological problem in the case of a highly excited spin system. Unfortunately, as  $S$  increases above unity (and also as  $T$  increases from zero), the number of dynamical equations increases rapidly, and the difficulties entailed in their solution or analysis increase even more rapidly. But there is reason to expect that the most important characteristics of the behavior of a highly anisotropic magnetic material with spin  $S > 1$  that fall outside the limits of “competence” of the magnetization vector can at least qualitatively be understood in an investigation with Eqs. (9a)–(9d). Perhaps an approach that has the generality of the phenomenological theory, and at the same time allows us to take account of the new nonvector degrees of freedom will also turn out to be useful.

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#### APPENDIX

As can be seen from (7) and (8), the single-site Hamiltonian  $\mathcal{H}_n$  (or  $\mathcal{H}'_n$ ) is time-dependent even if the total Hamiltonian  $\mathcal{H}$  does not vary in time. The expansion, customarily used in such cases,<sup>22</sup> of the wave function  $\Psi(t)$  in terms of the solutions to the quasistationary equation  $[\mathcal{H}'_n(t) - \epsilon_{n,i}(t)]\varphi_{n,i}(t) = 0$  requires that the fields entering into  $\mathcal{H}'_n$  vary slowly and no motion arise in the zeroth (adiabatic) approximation:  $d\langle \varphi_{n,0} | O_n | \varphi_{n,0} \rangle / dt = 0$ . We can get rid of these deficiencies by going over with the aid of a  $4S$ -parameter unitary transformation  $T_n(t)$  to the proper representation  $\Psi_n(t) = \exp[-i\mu(t)T]_n(t)\psi''_n$ , in which  $\psi''_n$  does not depend on  $t$ . In the new representation the equations of motion have the form

$$\langle \psi_n'' | [O_n'', \tilde{\mathcal{H}}_n'' + i\hbar \dot{T}_n^{-1} T_n] | \psi_n'' \rangle = 0 \quad (\text{A.1})$$

and are, for a given  $\psi''$ , equations for the parameters of  $T_n(t)$ . In view of the arbitrariness in  $O_n''$ , the function  $\psi_n''$  should be one of the eigenstates of the operator  $\tilde{\mathcal{H}}_n''^{\text{eff}} = \tilde{\mathcal{H}}_n'' + i\hbar \dot{T}_n^{-1} T_n$  that correspond to the energy  $\varepsilon = \dot{\mu}(t)$ . The choice of a specific function  $\psi_n''$  is determined by the initial conditions; by expanding in terms of the eigenfunctions of  $\tilde{\mathcal{H}}_n''^{\text{eff}}$ , we can show that the subsequent evolution of the solution to the complete Schrödinger equation is governed only by this function  $\psi_n''$ , and that it is, in this sense, adiabatic.

In the  $S = 1$  case the function  $\psi_n''$  can be chosen uniquely:  $\psi_n'' = |1\rangle$ . Separating out in  $T_n(t)$  the part that is connected only with space rotations, we have

$$\psi_n = T_n \psi_n'' = R_n \psi_n', \quad \psi_n' = \exp(-i2g_n Q_n^{xy}) \psi_n'',$$

and Eq. (A.1) in the new coordinate system has the form

$$\langle \psi_n' | [O_n', \tilde{\mathcal{H}}_n'^{\text{eff}}] | \psi_n' \rangle = 0, \quad (\text{A.2})$$

where

$$\tilde{\mathcal{H}}_n'^{\text{eff}} = \tilde{\mathcal{H}}_n' - \hbar \dot{\Phi}_n S_n' - \hbar 2\dot{g}_n Q_n^{\xi\eta}, \quad (\text{A.3})$$

so that  $\psi_n'$  is an eigenfunction of (A.3). The coincidence of  $\psi_n'$  with the function (4) and the coincidence of the new coordinate system with the MCS defined by the conditions (3) follow from the fact that the equations (A.2) are equivalent to Eqs. (9a)–(9d), a fact which can be verified directly, e.g.,

$$\langle \psi_n' | [S_n^{\xi}, 2\dot{g}_n Q_n^{\xi\eta}] | \psi_n' \rangle = i \frac{d}{dt} \langle \psi_n' | S_n^{\xi} | \psi_n' \rangle = i \dot{s}_n.$$

<sup>1</sup>The principal approximation made in the derivation of the LLE<sup>18</sup> consists in the transition from the  $(S_n^x)^2$ -type singlet-site operators to the magnetization functions  $[S^x(\mathbf{r})]^2$ , a process which corresponds to the semiclassical uncoupling process  $\langle (S_n^x)^2 \rangle \rightarrow \langle S_n^x \rangle^2$ .

<sup>2</sup>The description of the dynamics of a magnetic material with the aid of the density matrix does not constitute a difficulty formally. But for the problem formulated it is not more informative, and yet requires the consideration of the complete set of  $4S(S+1)$  variables connected by  $2S$  relations (in the absence of relaxation), which is not a simple problem even for an ensemble of noninteracting  $S = 1$  spins.<sup>19,20</sup> For  $S = 1/2$  both approaches lead to the vector LLE with the single relation  $|s| = \text{const}$ .

<sup>3</sup>The method of preliminary partial diagonalization of the single-site Hamiltonian  $\tilde{\mathcal{H}}_n$  allowed us earlier to find the analytic solutions to the self-consistent problem for ferromagnets and antiferromagnets possessing a symmetry plane.<sup>9,10</sup> The direct diagonalization for  $S = 1$ , on the other hand, requires the solution of the complete set of six equations. This is a much more difficult problem, and has as yet not been solved analytically even in the real-matrix case, which requires only three rotations.<sup>17</sup>

<sup>4</sup>The possibility of such a vortex follows from symmetry arguments, and was first pointed out by Salomaa and Volovik<sup>27</sup> without reference to the assumptions about the spin-contraction mechanism.

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