

$\Delta T'$  for single-crystal  $\text{CdCr}_2\text{Se}_4$  (sample I), in which no selenium deficiency is detected, and for single-crystal gadolinium; both values are close to  $4^\circ$ . A similar investigation was made of two  $\text{CdCr}_2\text{Se}_4$  single crystals having a selenium deficiency up to 1.5%. It turned out that  $\Delta T'_c = 26^\circ$  for one of them (sample II) and  $\Delta T'_c = 35^\circ$  for the other (sample III). After the described annealing of sample III in the selenium atmosphere, the value of  $\Delta T'_c$  decreased to  $12^\circ$ . We conclude therefore that the shift  $\Delta T'_c$  is due to the selenium deficiency of the sample and to the impurity.

The foregoing examples show clearly that the Curie point of magnetic semiconductors must be determined by methods that exclude the external magnetic field, since the error can be quite large.

In conclusion, we thank É. L. Nagaev and A. P. Grigin for a discussion of the results.

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## Bound states of a large number of magnons in a ferromagnet with a single-ion anisotropy

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(Submitted December 28, 1976)  
Zh. Eksp. Teor. Fiz. 72, 2000–2015 (May 1977)*

The conditions for the formation of a magnon bound state in one- and three-dimensional ferromagnets with the "easy axis" type of anisotropy are investigated. Explicit expressions are obtained in the one-dimensional case for the energies and wave functions of the bound states of an arbitrary number of magnons under the natural assumption that the magnetic anisotropy energy is small compared to the exchange energy. The binding energy per magnon increases monotonically with increasing number of magnons. For large numbers of magnons ( $N \gg 1$ ) the solution to the quantum problem goes over into the specific self-localized solution to the Landau-Lifshitz equation for the magnetization vector. It is shown that, if there do not exist in a three-dimensional ferromagnet bound states of two magnons with a low quasimomentum, then there can arise in this ferromagnet bound states of a large number,  $N$ , of magnons (in the case when  $N \geq N_c$ , where the quantity  $N_c$  is determined by the ratio of the exchange energy to the anisotropy energy). The form of the self-localized solution is obtained by means of a numerical solution of the nonlinear equation of motion for the magnetization vector, and the dependence of the parameters of this solution on  $N$  is determined. An interpretation is given of the physical meaning of the bound state of a large number of magnons.

PACS numbers: 75.30.Gw

### INTRODUCTION

The physical properties of magnetically ordered crystals at low temperatures are determined to a considerable extent by the properties of the gas of elementary

excitations of the magnetic substance—the magnons. The weakly excited states of a magnetic substance are usually described in terms of an ideal quasiparticle gas, i. e., in the approximation of noninteracting magnons. It is, however, clear that the interaction of the magnons

can lead to a substantial reconstruction of the elementary-excitation spectrum, in particular, to the formation of magnon bound states. Since magnons are very similar in their statistical properties to Bose particles, which are capable of "agglomerating" in the low-energy states, the presence of magnon bound states can lead to the spatial inhomogeneity of the excited state of a ferromagnet.

A substantial portion of the investigations of magnon bound states pertains to one-dimensional magnetic substances. Interest in the one-dimensional models is due both to the appreciable simplification of their analysis (while retaining a number of important physical features of the general problem) and to the fact that some real magnetic substances are well described by quasi-one-dimensional models. The problem of the existence of bound states of an arbitrary number of magnons in an isotropic Heisenberg chain with spin  $\frac{1}{2}$  was first formulated and fully analyzed by Bethe.<sup>[1]</sup> In a number of subsequent papers<sup>[2-4]</sup> Bethe's results were generalized to the case of an anisotropic Heisenberg chain.

In the present paper we consider magnon bound states in one- and three-dimensional ferromagnets with an arbitrary spin value at the lattice sites in the presence of a single-ion anisotropy. In the spin variables the anisotropy energy  $\mathcal{H}_A$  can be represented in the form

$$\mathcal{H}_A = -\frac{\beta\mu_0 M_0}{s} \sum_l s_{zl}^2 \quad (1)$$

where  $s_l$  is the spin operator at the  $l$ -th site ( $s$  is the magnitude of the spin),  $\beta$  is the magnetic anisotropy constant ( $\beta > 0$ ),  $\mu_0$  is the Bohr magneton, and  $M_0 = 2\mu_0 s/a^3$  is the saturation magnetization ( $a^3$  is the volume of the unit cell of the crystal).

We succeeded in deriving analytic expressions for the energies of the bound states of  $N$  magnons in the one-dimensional case under the natural assumption that the magnetic-anisotropy energy is significantly smaller than the exchange energy (i.e., that  $\beta\mu_0 M_0 \ll I$ , where  $I$  is the ferromagnet's exchange integral, equal in order of magnitude to the Curie temperature). In describing the excited states of a ferromagnetic chain with  $N \ll N_1$ , where  $N_1 = 4s(I/2\mu_0\beta M_0)^{1/2} \gg 1$ , we used the Holstein-Primakoff representation, and reduced the problem of the  $N$ -magnon bound state to that of the ground state of a one-dimensional system of a finite number of bosons with a  $\delta$ -function pair attraction. Such a problem admits of an exact solution.<sup>[5]</sup>

It is to be expected that we can use in the investigation of states with a large number of spin deviations the classical description in terms of the macroscopic magnetization density  $\mathbf{M}(\mathbf{r}, t)$ , for which equations of motion (the Landau-Lifshitz equations) exist. The energy,  $W\{\mathbf{M}\}$ , of the ferromagnet and the number,  $N\{\mathbf{M}\}$ , of spin deviations are functionals of the magnetization  $\mathbf{M}(\mathbf{r}, t)$ . The magnetization distribution corresponding to the bound state of a large number of magnons, is realized by the minimum of the energy functional  $W\{\mathbf{M}\}$  for a given integral value of  $N\{\mathbf{M}\}$ . This distribution corre-

sponds to the self-localized solution to the nonlinear Landau-Lifshitz equation for the magnetization.

We have verified that for  $1 \ll N \ll N_1$  the quantum and classical expressions for the bound-state energy and for the spatial magnon density coincide. Thus, the analysis of the one-dimensional model allowed us to draw the conclusion that the bound state of a large number ( $N \gg 1$ ) of magnons in a ferromagnet is quasiclassical in nature and, thereby, to significantly simplify the analysis of the conditions for the existence of  $N$ -magnon bound states in the three-dimensional case. It is well known that, in contrast to the one-dimensional case, two quasiparticles will form a bound state in a three-dimensional crystal only when the amplitude of the attractive interaction between them exceeds some critical value. In an isotropic Heisenberg ferromagnet, this circumstance leads to the absence of bound states of two magnons with a small total quasimomentum.<sup>[6]</sup> As shown in Ref. 7, the presence of magnetic anisotropy leads to the appearance of bound states of two magnons with a small total quasimomentum only when  $\mu_0\beta M_0 \gtrsim I$ , i.e., only upon the fulfillment of conditions that are far from reality. We shall show that the condition for the existence of a bound state of a large number,  $N$ , of magnons with zero total quasimomentum assumes another form and becomes less rigid. To wit, an  $N$ -magnon bound state is formed when

$$N^{2/3} \mu_0 \beta M_0 \gg I.$$

At the end of the paper we shall discuss the physical meaning of the bound state of a large number of magnons, interpreting it as a magnon drop.<sup>[8]</sup>

## § 1. QUANTUM ANALYSIS OF THE BOUND STATE OF A FINITE NUMBER OF MAGNONS IN A FERROMAGNETIC CHAIN

A standard procedure for an approximate quantum analysis of a weakly excited state of a ferromagnet consists in the use of the Holstein-Primakoff representation, which allows us to go over from the spin operators to the magnon creation and annihilation operators  $a^+$  and  $a$ . In terms of these operators we can write

$$s_z(l) = s - a_l^+ a_l, \quad (2)$$

where  $a^+(l) a(l)$  has the meaning of a number operator for the spin deviations at the site  $l$  and  $s$  is the value of the atomic spin. When the number of excitations is not large, the Hamiltonian of the magnon gas in the  $k$ -representation is determined by the expression

$$\mathcal{H} = \sum_k \varepsilon(k) a_k^+ a_k + \mathcal{H}_1, \quad (3)$$

where  $\varepsilon(k)$  is the energy of the one-magnon state ( $a_k^+ a_k$  is the number operator for the magnons in the state  $k$ ), while the operator  $\mathcal{H}_1$  describes the magnon interaction.

The dominant magnon interaction stems, generally speaking, from the exchange interaction. It is well known, however, that the exchange-interaction-produced two-magnon scattering amplitude is proportional

to  $I(ak_1)(ak_2)$ , where  $k_1$  and  $k_2$  are the wave vectors of the colliding spin waves. Therefore, in the long-wave approximation ( $ak \rightarrow 0$ ), the interaction of exchange origin becomes vanishingly small. At the same time, owing to the single-ion anisotropy energy (1), the magnon interaction gives a nonzero spin-wave scattering amplitude even in the limit as  $k \rightarrow 0$ . Indeed, if we substitute (2) into (1) and limit ourselves to only terms of fourth order in the operators  $a_k^+$  and  $a_k$ , then we can easily derive for the Hamiltonian of the magnon-magnon interaction the expression<sup>1)</sup>

$$\mathcal{H}_1 = \frac{1}{L} \sum V(k_1 k_2, k_3 k_4) a_1^+ a_2^+ a_3 a_4 \Delta(k_1 + k_2 - k_3 - k_4), \quad (4)$$

where  $\Delta(k)$  is the Kronecker symbol,  $L$  is the length of the chain, and the amplitude of the interaction possesses the following limit for  $k \rightarrow 0$ :

$$V \rightarrow -V_0 = -2\mu_0^2 \beta. \quad (5)$$

The principal characteristic of the magnon interaction in a ferromagnet with an "easy-axis" type of anisotropy ( $\beta > 0$ ) consists in such a behavior of the amplitude  $V(k_1 k_2, k_3 k_4)$ .

Taking interest henceforth in only the long-wave approximation ( $ak \ll 1$ ), we can restrict ourselves to the consideration of only the contact interaction of the magnons and reduce the expression for  $\mathcal{H}_1$  as follows:

$$\mathcal{H}_1 = -\frac{V_0}{L} \sum a_1^+ a_2^+ a_3 a_4 \Delta(k_1 + k_2 - k_3 - k_4). \quad (6)$$

It is clear that in the case of such a simplification of the Hamiltonian, we are justified in describing only those inhomogeneous states of the magnetic substance for which the characteristic magnon wave vector satisfies the condition

$$I(ak)^2 \ll \mu_0 \beta M_0. \quad (7)$$

Assuming that the condition (7) is fulfilled, let us rewrite the Hamiltonian, (3), of the ferromagnet in the long-wave approximation:

$$\mathcal{H} = \sum_n [\varepsilon_0 + I(ak)^2] a_n^+ a_n - \frac{V_0}{L} \sum_n a_n^+ a_n^+ a_n a_n \Delta(k_1 + k_2 - k_3 - k_4), \quad (8)$$

where  $\varepsilon_0 = \hbar\omega_0 = 2\mu_0 \beta M_0$  is the frequency of the homogeneous ferromagnetic resonance and  $I$  coincides in order of magnitude with the exchange integral.

In the coordinate representation, a magnon-magnon interaction of the form (6) is described by the potential energy

$$\mathcal{H}_1 = \sum_{i < j} U(x_i - x_j), \quad U(x) = -aV_0 \delta(x), \quad (9)$$

where  $x_i$  is the coordinate of the  $i$ -th quasiparticle. The problem of the bound state of  $N$  bosons interacting according to the law (9) admits of an exact solution.<sup>[5]</sup> This state is described by a wave function of the form

$$\Phi(x_1 \dots x_N) = \exp \left\{ -\frac{\mu_0 \beta M_0}{Ia} \sum_{i < j} |x_i - x_j| \right\}, \quad (10)$$

while the bound-state energy  $E(N)$  is determined by the formula

$$E(N) = N\varepsilon_0 \left( 1 - \frac{N^2 - 1}{3N^2} \right), \quad N_1 = 4s \left( \frac{I}{2\mu_0 \beta M_0} \right)^{1/2}. \quad (11)$$

We shall assume that the number  $N_1$  is large ( $N_1 \gg 1$ ). Using the explicit form of the wave function (10), we can easily derive an expression for the mean wave vector of the magnons forming the bound state. It turns out that for  $N \gg 1$  we have in order magnitude

$$I(ak)^2 \approx (N/N_1)^2 \mu_0 \beta M_0. \quad (12)$$

Comparing (12) with the condition (7), we see that the Hamiltonian (8) can be used only to describe magnon bound states with  $N \ll N_1$ . Thus, the assumption that  $N_1 \gg 1$ , which is equivalent to the assumption that the magnetic-anisotropy energy is small compared to the exchange energy (i. e., that  $\mu_0 \beta M_0 \ll I$ ), is extremely important. In terms of the one-magnon excitations, the latter inequality corresponds to the smallness of the energy of interaction of two quasiparticles in comparison with the energy-band width of the free quasiparticles. When this inequality is fulfilled, the localization radius of the bound state is much greater than the range of the two-body forces, which completely justifies the replacement of the interaction potential by the  $\delta$ -function (9). Notice that, when this condition is fulfilled, two-quasiparticle bound states are not formed in a three-dimensional crystal.

## § 2. QUASICLASSICAL DESCRIPTION OF THE BOUND STATE OF A LARGE NUMBER OF MAGNONS. THE ONE-DIMENSIONAL CASE

We have convinced ourselves that the quantum analysis of the magnon bound states on the basis of the Hamiltonian (8) makes sense only when  $N \ll N_1$ . But if  $N \gg 1$ , then the description of the bound states of a large number,  $N \gtrsim N_1$ , of magnons can be approached from another angle. It was recently shown<sup>[9]</sup> that the bound state of a large number of phonons in an anharmonic chain is the quantum analog of the classical self-localized vibrations of a nonlinear string, which are describable in terms of the macroscopic equations of vibrations.<sup>[10]</sup> A similar situation should obtain in a magnetic substance to which the quantum model used by us in the preceding section, as well as the macroscopic description in terms of the nonuniform magnetization density, is applicable. It is well known that the classical behavior of a magnetic substance can be studied with the aid of the Landau-Lifshitz equations.<sup>[11]</sup> Therefore, it is natural to raise the question of the existence of self-localized solutions of the one-dimensional nonlinear Landau-Lifshitz equation and of the correspondence of these solutions to the bound states of a large number of magnons.

Let  $M(r)$  be the density of the magnetization vector of the ferromagnet. The presence of self-localized oscillations implies that in a bounded region of space (in the one-dimensional case, in a finite section of the chain) the magnetization,  $M_z(r)$ , along the easy axis differs from the nominal value  $M_0$ , and there arises a fi-

nite total deviation of the  $z$  component of the magnetic moment from the equilibrium value. Let us define the number,  $N$ , of magnons in such a state as the total number of spin deviations in the system, to wit,

$$N = \frac{1}{2\mu_0} \int [M_0 - M_z(r)] dr. \quad (13)$$

Since the Hamiltonian, (4), of the interaction commutes with the total  $z$  component of the magnetic moment, the total number of magnons is an integral of the motion in both the quantum and classical descriptions.

The requirement that the quantity  $N(N \gg 1)$  in the classical relation (13) should have integral values is equivalent to the quasiclassical quantization of the corresponding localized solutions of the Landau-Lifshitz equation. The Landau-Lifshitz equations can be derived with the aid of a variational principle which uses the expression for the energy of the ferromagnet and which is written in the form of a functional of the magnetization vector  $M(\mathbf{r})$ . The energy,  $W$ , of the ferromagnet includes the exchange energy, the magnetic-anisotropy energy, and the energy of the magnetic-dipole interaction. We shall henceforth assume that  $\beta \gg 4\pi$ . When this inequality is fulfilled, the contribution of the dipole interactions to the energy of the ferromagnet is small,<sup>2)</sup> and allowance for them insignificantly (to the extent of  $4\pi/\beta$ ) alters the form of the self-localized solutions of the Landau-Lifshitz equations of interest to us. Therefore, we shall restrict ourselves to the consideration of the exchange-interaction energy and the anisotropy energy (1). Then

$$W\{\mathbf{M}\} = \frac{1}{2} \int \left\{ \alpha \left( \frac{\partial \mathbf{M}}{\partial r_i} \right)^2 - \beta M_z^2 \right\} dr, \quad (14)$$

where  $\alpha = Ia^2/2\mu_0 M_0$  is the exchange constant. The equations for the free motion of the magnetization, which are the Landau-Lifshitz equations with relaxation neglected, can be written in the form

$$\frac{\partial \mathbf{M}}{\partial t} = \frac{2\mu_0}{\hbar} [\mathbf{H}_{\text{eff}}, \mathbf{M}], \quad \mathbf{H}_{\text{eff}} = -\frac{\delta W\{\mathbf{M}\}}{\delta \mathbf{M}}. \quad (15)$$

We shall be interested in the localized solutions to (15), i. e., in the solutions for which  $M(\mathbf{r}) \rightarrow M_0$  as  $|\mathbf{r}| \rightarrow \infty$ . Let us choose the  $z$  axis along  $M_0$ , and let us represent the components of the density of the magnetization vector in the form

$$M_x = M_0 \cos \theta, \quad M_x + iM_y = M_0 \sin \theta e^{i\varphi}. \quad (16)$$

Let us consider the localized solutions for which

$$\theta = \theta(r), \quad \varphi = \omega t, \quad \omega = \text{const}, \quad (17)$$

where the angle  $\theta(r) \rightarrow 0$  as  $|\mathbf{r}| \rightarrow \infty$ . The sought solutions describe nonuniform, localized precession of the magnetization with frequency  $\omega$  and a coordinate-dependent amplitude. We shall clarify the quantum meaning of the quantity  $\omega$  below. Equation (15) with allowance for (16) and (17) assumes the form

$$\frac{\alpha}{\beta} \Delta \theta - \sin \theta \cos \theta + \frac{\omega}{\omega_0} \sin \theta = 0. \quad (18)$$

Returning to the one-dimensional problem, and introducing the coordinate  $\xi$  along the chain,<sup>3)</sup> we rewrite Eq. (18) in the form:

$$\frac{\alpha}{\beta} \frac{d^2 \theta}{d\xi^2} - \sin \theta \cos \theta + \frac{\omega}{\omega_0} \sin \theta = 0. \quad (19)$$

The localized solution to Eq. (19) that decreases monotonically as  $\xi \rightarrow \pm \infty$  can be written in terms of elementary functions, to wit:

$$\text{tg} \frac{\theta}{2} = \left( \frac{\omega_0 - \omega}{\omega} \right)^{1/2} \text{sech} \left\{ \left[ \frac{\beta}{\alpha} \left( \frac{\omega_0 - \omega}{\omega_0} \right) \right]^{1/2} (\xi - \xi_0) \right\}, \quad \xi_0 = \text{const}. \quad (20)$$

We see that the amplitude and frequency,  $\omega$ , of the self-localized oscillation are connected by a simple functional dependence. Substituting (20) into the expression (14), we find the energy of the self-localized oscillation<sup>4)</sup> as a function of the frequency  $\omega$ :

$$E = 4a^2 M_0^2 (\alpha\beta)^{1/2} \left( \frac{\omega_0 - \omega}{\omega_0} \right)^{1/2}. \quad (21)$$

Using (20) and the condition (13), we also find the number,  $N$ , of spin deviations that corresponds to the self-localized oscillation of frequency  $\omega$ :

$$N = N_1 \text{ arch} (\omega_0/\omega)^{1/2}, \quad (22a)$$

or

$$\omega = \frac{\omega_0}{\text{ch}^2(N/N_1)}. \quad (22b)$$

The relations (22) establish a connection between the frequency  $\omega$  and the number of magnons forming the self-localized state. Now we can express the energy of a quasiclassical bound state of a large number of magnons in terms of the number of magnons:

$$E(N) = \varepsilon_0 N_1 \text{ th} (N/N_1). \quad (23)$$

A plot of the dependence  $E = E(N)$  is shown in Fig. 1, while a schematic plot of  $\theta = \theta(\xi)$  for  $N \gg N_1$  is given in Fig. 2.

For  $N \gg N_1$ , it follows from (23) that

$$E = \varepsilon_0 N_1 = 4a^2 M_0^2 \sqrt{\alpha\beta}. \quad (24)$$

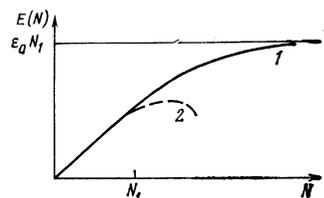


FIG. 1. The bound-state energy as a function of the magnon number  $N$  (the one-dimensional case): 1) the quasiclassical dependence  $E(N)$  determined by the formula (23); 2) extrapolation of the dependence (11) to values of  $N \sim N_1$ .

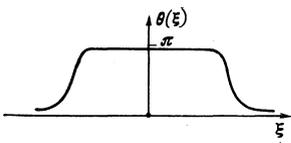


FIG. 2. Schematic representation of the magnetization distribution in self-localized oscillations when  $\omega \ll \omega_0$  (the one-dimensional case).

It is easy to verify that (24) coincides with twice the energy of a  $180^\circ$  domain wall (for an area of  $a^2$  of the domain boundary). This is an entirely natural result, since, for  $N \gg N_1$ , the whole inhomogeneity of the magnetization distribution is practically concentrated in the transition regions for the magnetization (see Fig. 2). Outside the narrow regions of width  $\Delta\xi \sim (\alpha/\beta)^{1/2}$  each magnetization intensity corresponds to the nominal value, and differs only in direction outside and inside the region of localization of the deviation of the magnetization from its value at infinity.

For  $N \ll N_1$  we have

$$E(N) = \varepsilon_0 N (1 - N^2/3N_1^2). \quad (25)$$

Comparing (25) with (11), we see that the quasiclassical expression for the  $N$ -magnon bound state energy in the case when  $1 \ll N \ll N_1$  coincides with the corresponding quantum expression to within  $1/N^2$ . Notice that the classical analog of the bound state can be found directly by minimizing the energy functional, (14), of the magnetic substance for a given value of the magnon-number functional (13), namely, from the condition

$$\delta[W\{M\} - \hbar\omega N\{M\}] = 0, \quad (26)$$

where  $\hbar\omega$  is a Lagrange multiplier. The Euler-Lagrange equation arising from (26) coincides with Eq. (18), i. e., the magnetization distribution minimizing the energy of the magnetic substance for a given finite value of the total number of magnons is a self-localized solution of the nonlinear Landau-Lifshitz equation.

Using (26), we can easily derive in a general form an important relation that uncovers the quantum meaning of the frequency of precession of the magnetization in the self-localized state:

$$\hbar\omega = dE(N)/dN. \quad (27)$$

This relation is equivalent to the assertion that, when the number of magnons in the bound state is increased by one, the energy of such a state increases by  $\hbar\omega$ . Consequently, the quantity  $\hbar\omega$  is the energy of excitation of one reversed spin in the ferromagnet containing the bound state of a large number,  $N$ , of magnons.

### § 3. BOUND STATES OF A LARGE NUMBER OF MAGNONS IN A THREE-DIMENSIONAL FERROMAGNET

As was shown in the preceding section, bound states of a large number of magnons ( $N \gg 1$ ) can be obtained

by means of a quasiclassical quantization of the self-localized solutions to the Landau-Lifshitz equation for the magnetization. Below we shall convince ourselves that, in a three-dimensional ferromagnet with an easy-axis type of anisotropy,  $N$ -magnon bound states can form only when  $N \gg 1$ ; therefore, we shall investigate them, using the quasiclassical method developed in the preceding section.

It is to be expected that, without allowance for the magnetic-dipole interaction, the lowest energy is possessed in a three-dimensional ferromagnet by the centrally symmetric solution  $\theta = \theta(r)$ . The equation for  $\theta(r)$  is obtained directly from (18):

$$\frac{\alpha}{\beta} \left( \frac{d^2\theta}{dr^2} + \frac{2}{r} \frac{d\theta}{dr} \right) - \sin\theta \cos\theta + \frac{\omega}{\omega_0} \sin\theta = 0. \quad (28)$$

Although we can integrate Eq. (28) only numerically, the question of the existence of self-localized solutions of this equation can be qualitatively analyzed virtually to the end without recourse to the explicit form of the solutions. This can be done, using the phase-plane method.

Let us introduce the notation  $\theta' = d\theta/dr$  and consider the phase plane  $(\theta, \theta')$ . It is convenient to study the behavior of the integral curves (trajectories) in this plane by the standard method,<sup>[12]</sup> introducing the function  $C(r)$ , which is the first integral of motion for the "reduced" equation, obtained from (28) by eliminating the term with the first derivative. This "reduced" equation coincides with (19) if we set  $\xi = r$ . The function  $C(r)$  has the form

$$C(r) = \frac{1}{2} \left( \frac{\alpha}{\beta} \right) \left( \frac{d\theta}{dr} \right)^2 + \frac{1}{2} \cos^2\theta - \frac{\omega}{\omega_0} \cos\theta. \quad (29)$$

It is easy to verify that  $C(r)$  is a monotonically decreasing function of the coordinate  $r$ :

$$\frac{dC}{dr} = -\frac{\alpha}{\beta} \frac{2}{r} \left( \frac{d\theta}{dr} \right)^2, \quad (30)$$

i. e., decreases with increasing  $r$ .

To carry out a qualitative analysis of the solutions to Eq. (28), let us depict in the phase plane curves of constant level of the quantity  $C$ , conditionally calling them constant-energy curves.<sup>5)</sup> Typical constant-energy curves for  $\omega < \omega_0$  are shown in Fig. 3. It should only be kept in mind that, by definition, the polar angle  $\theta$  varies

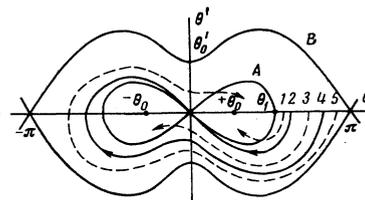


FIG. 3. Schematic drawing of the phase trajectories in the  $(\theta', \theta)$  plane. The points  $\pm\theta_0$  are foci; the curves A and B are the separatrices for the integral curves of the "reduced" equation.

in the interval  $0 \leq \theta < \pi$ . In allowing negative values of  $\theta$  in the phase plane, we should remember that the transition from the positive semiaxis ( $\theta > 0$ ) to the negative semiaxis ( $\theta < 0$ ) is connected with a change in the phase of the precession by a half-period (by the quantity  $\pi/\omega$ ). The points  $(\pm \theta_0, 0)$ , where  $\cos \theta_0 = \omega/\omega_0$ , are foci corresponding to  $C = -\omega^2/2\omega_0^2$  and describing the state of uniform precession of the magnetic substance with frequency  $\omega$ . The passage to these limiting states cannot be of interest to us for two reasons: first, they are not localized ( $\theta = \theta_0 = \text{const} \neq 0$ ) and, secondly, they are mechanically unstable. The curve A corresponds to  $C = C_A = \frac{1}{2} - \omega/\omega_0$  and passes through the coordinate origin and the points  $(\pm \theta_1, 0)$ , where  $\cos \theta_1 = 2\omega/\omega_0 - 1$ . The curve B corresponds to the value  $C = C_B = \frac{1}{2} + \omega/\omega_0$  and passes through the point  $\theta = \pi$  on the abscissa axis and the points  $\theta' = \theta'_0 = \pm 2(\omega/\omega_0)^{1/2}$  on the ordinate axis. The curves A and B are, in fact, separatrices, dividing the phase plane into three regions. In the first region ( $C < C_A$ ), i. e., inside the separatrices, are located curves that close round one of the foci. In the second region ( $C_A < C < C_B$ ), i. e., between the separatrices, pass constant-energy curves enclosing both foci. To the third region ( $C > C_B$ ) belong constant-energy curves that do not intersect the  $\theta' = 0$  axis—they will not be investigated below.

The phase trajectories of Eq. (28) intersect the constant-energy curves, since along them the quantity  $C$  decreases with increasing  $r$ .

We shall seek the self-localized solutions of Eq. (28) that are bounded at  $r=0$ . These solutions automatically satisfy the condition  $d\theta/dr = 0$  at  $r=0$  (the starting point of the integral curve lies on the  $\theta' = 0$  axis). For definiteness, let us assume that  $\theta > 0$  at  $r=0$ .

It is clear that the phase trajectories that start from inside the separatrix A terminate, for  $r \rightarrow \infty$ , at the focus  $\theta = +\theta_0$  and describe nonlocalized, unstable states. Consequently, the beginning,  $\theta = \theta(0)$ , of a phase trajectory describing a localized state should lie at least in the interval  $\theta_1 < \theta(0) < \pi$  on the abscissa axis. As a rule, the phase trajectories that have their origins in this interval intersect the separatrix A, and therefore "drop out of the game," terminating at one of the foci  $\theta = \pm \theta_0$  (the dashed curves 1, 3, and 5 in Fig. 3). However, there exist, besides this manifold of phase trajectories, curves which correspond to a discrete set of  $\theta(0)$  values and which terminate at the coordinate origin as  $r \rightarrow \infty$  (the curves 2 and 4 in Fig. 3). These selected trajectories depict the solutions to Eq. (28) that vanish at infinity, i. e., the localized states of interest to us.

The asymptotic behavior of the function  $\theta = \theta(r)$  for the localized states is easily obtained by linearizing Eq. (28) for  $\theta \ll 1$ . It turns out that for these solutions we have for  $r \rightarrow \infty$

$$\theta(r) = \frac{\text{const}}{r} \exp \left\{ -r \left[ \frac{\beta}{\alpha} \left( 1 - \frac{\omega}{\omega_0} \right) \right]^{1/2} \right\}. \quad (31)$$

It is natural to classify the localized solutions according to the number of zeroes (nodes), i. e., according to the number of intersections of the  $\theta' = 0$  axis by the

phase trajectory. To the "ground" state corresponds the monotonically decreasing function  $\theta(r)$  whose phase trajectory is represented by the curve 2 in Fig. 3. The subsequent analysis will be devoted to the elucidation of the characteristic features of the "ground" state.

We shall first of all study the self-localized ground state for  $\omega \ll \omega_0$ . For  $\omega \ll \omega_0$  the values of  $C_A$  and  $C_B$  differ little:  $C_B - C_A = 2\omega/\omega_0 \ll C_A$ , and the separatrices A and B almost coincide (in Fig. 3 the extreme points of the separatrices are characterized by the quantities  $\theta_1 = \pi - 2\sqrt{\omega/\omega_0}$  and  $\theta'_0 = 2\sqrt{\omega/\omega_0}$ ). Consequently,  $\theta(0)$  lies in a narrow interval  $\Delta\theta \sim 2\sqrt{\omega/\omega_0}$  near  $\theta = \pi$ . Since the derivative  $d\theta/dr$  near the point  $\theta = \pi$  is small, there exists a sufficiently wide range of variation of the variable  $r$  in which  $\theta(r)$  virtually does not differ from  $\pi$ :

$$\theta(r) - \theta(0) = -\frac{1}{6} [\pi - \theta(0)] \left( \frac{\beta}{\alpha} \right) r^2. \quad (32)$$

Since  $\pi - \theta(0) < 2\sqrt{\omega/\omega_0}$ , it follows from (32) that we can assume  $\theta(r) \approx \theta(0) \approx \pi$  right up to  $r \sim l = \sqrt{\alpha/\beta}$  (as we shall verify below, this estimate significantly reduces the dimension of the region in which  $|\theta(r) - \pi| \ll 1$ ). The behavior of the function  $\theta(r)$  for  $r \gg \sqrt{\alpha/\beta}$  in the  $\omega \ll \omega_0$  case is also easy to establish. Indeed, in this case the characteristic length parameter,  $l$ , determining the order of magnitude of the derivatives of the function  $\theta(r)$  ( $l^2 = \alpha/\beta$ ) is small compared to  $r$ . Therefore, the term with the first derivative in (28) is small compared to the second derivative, and Eq. (28) for  $r \gg l$  and  $\omega \ll \omega_0$  reduces to the one-dimensional equation:

$$\frac{\alpha}{\beta} \frac{d^2\theta}{dr^2} = \sin \theta \cos \theta. \quad (33)$$

The solution to Eq. (33) that vanishes at infinity ( $r \rightarrow \infty$ ) is known:

$$\cos \theta(r) = \text{th} \left[ (\beta/\alpha)^{1/4} (r-R) \right], \quad R = \text{const}. \quad (34)$$

Notice that this solution describes the magnetization distribution in the plane of a domain wall in the ferromagnet. Thus, for  $\omega \ll \omega_0$  the localized solution describes the following state of the magnetic substance. In the macroscopic region of the radius  $R \gg l$ , the magnetization corresponds to the nominal magnetization, but with a direction opposite to that of the vector  $\mathbf{M}$  ( $\theta \approx \pi$ ). This region is separated from the remaining magnetic substance by a spherical "domain boundary" of radius  $R$  and thickness  $l$ , in which the magnetization executes precession with frequency  $\omega$  and angle  $\theta$  that depends on the coordinate  $r$ . Virtually all the energy of such a state is concentrated in a transition region of thickness  $l$ , and coincides with the energy of the spherical domain boundary:

$$W = 4\pi R^2 \cdot 2\beta l M_0^2. \quad (35a)$$

The number of reversed spins in such a state is just as easy to determine if we consider only the volume part of it:

$$N = \frac{4\pi}{3} R^3 \left( \frac{M_0}{\mu_0} \right). \quad (35b)$$

Using (35a) and (35b), as well as the condition (26), we can connect the radius of the self-localized solution with the quantity  $\omega$ :

$$R(\omega) = \frac{2\omega_0}{\omega} \left( \frac{\alpha}{\beta} \right)^{1/2} \gg \left( \frac{\alpha}{\beta} \right)^{1/2} = l.$$

It is now easy to derive for  $\omega \rightarrow 0$  the following asymptotically exact expressions connecting the energy,  $E$ , of the self-localized oscillation with the number,  $N$ , of spin deviations:

$$E = 3\varepsilon_0 N (2N_3/3N)^{1/2}, \quad (36)$$

as well as the precession frequency with the number  $N$ :

$$\omega = 2\omega_0 (2N_3/3N)^{1/2}, \quad (37)$$

where

$$N_3 = 4\pi s (I/\hbar\omega_0)^{3/2} = 4\pi s (I/2\mu_0\beta M_0)^{3/2} \gg 1. \quad (38)$$

(Running ahead, let us note that the numerical analysis confirmed the validity of the relations (36) and (37) for  $\omega \ll \omega_0$ .)

Comparing the expressions (36) and (37), and taking (27) into account, we can see that the energy of excitation of one spin deviation going over into the "ground" bound state of  $N$  magnons is smaller than the energy per magnon:

$$\hbar\omega = \varepsilon_0 (E/N).$$

The condition  $\omega \ll \omega_0$  corresponds to the requirement that  $N \gg N_3$ , under which  $E \ll \varepsilon_0 N$ .

It is also possible to analyze the properties of the self-localized solution in another limiting case, namely, in the case when  $\omega_0 - \omega \ll \omega_0$ . If the frequency  $\omega$  is very close to  $\omega_0$ , then the separatrix A presses itself toward the coordinate origin ( $\theta_1 \ll 1$ ). But the "ground" state's trajectory, which does not intersect the  $\theta' = 0$  axis, should have its origin near the separatrix A, and therefore we should expect that, for it,  $\theta(r) \ll 1$ .

Assuming that the angle  $\theta$  is small, we can easily derive for it the equation

$$\frac{\alpha}{\beta} \Delta\theta - \left( 1 - \frac{\omega}{\omega_0} \right) \theta + \frac{1}{2} \theta^3 = 0.$$

Let us introduce a new scale for the space coordinates, introducing the dimensionless variable

$$x = \frac{r}{l} \left( 1 - \frac{\omega}{\omega_0} \right)^{1/2}, \quad l^2 = \frac{\alpha}{\beta}, \quad (39)$$

and let us define a new sought function  $\Psi(x)$ :

$$\theta = [2(1-\omega/\omega_0)]^{1/2} \Psi(x). \quad (40)$$

Then for  $\Psi$  we obtain the nonlinear equation

$$\frac{d^2\Psi}{dx^2} + \frac{2}{x} \frac{d\Psi}{dx} - \Psi + \Psi^3 = 0, \quad (41)$$

the properties of which have been well studied.<sup>[13-15]</sup>

The fundamental self-localized solution of Eq. (41) possesses an "amplitude"  $\Psi(0) \cong 4.34$ , and falls off over distances  $\Delta x \sim 1$ . Consequently,

$$\theta(0) = [2(1-\omega/\omega_0)]^{1/2} \Psi(0) \approx 6[1-\omega/\omega_0]^{1/2} \ll 1,$$

which justifies the initial assumption that  $\theta(r) \ll 1$ . Further, the the scaling transformation (39) shows that the self-localized state has a region of localization

$$\Delta r \sim l(1-\omega/\omega_0)^{-1/2} \gg l,$$

i. e., is very highly "smeared out" in space.

Let us now write down in that approximation in which (38) for  $\omega \rightarrow \omega_0$  is valid the bound-state energy

$$E = \frac{M_0^2}{2} \int \left\{ \alpha (\nabla\theta)^2 - \frac{1}{4} \beta \theta^4 \right\} dV + \frac{1}{2} \beta M_0^2 \int \left\{ \theta^2 - \frac{1}{12} \theta^4 \right\} dV \quad (42)$$

and the number of bound magnons

$$N = \frac{M_0}{4\mu_0} \int \left( \theta^2 - \frac{\theta^4}{12} \right) dV. \quad (43)$$

Comparing (42) and (43), and also taking into consideration (39) and (40), we obtain

$$E = N\varepsilon_0 + N_3\varepsilon_0(1-\omega/\omega_0)^{1/2} J_2, \quad (44)$$

where the constant  $J_2$  is equal to

$$J_2 = \frac{1}{4\pi} \int \left\{ \left( \frac{d\Psi}{dx} \right)^2 - \frac{1}{2} \Psi^4 \right\} dx,$$

while  $N_3$  is determined as before by the formula (38), and, similarly,

$$(1-\omega/\omega_0)^{1/2} = N_3 J_1 / N, \quad (45)$$

where the constant  $J_1$  is equal to

$$J_1 = \frac{1}{8\pi} \int \Psi^2 dx.$$

Combining (44) and (45), we can derive the asymptotic dependence of the bound-state energy on  $N$  for  $\omega \rightarrow \omega_0$ :

$$E = N\varepsilon_0 + N_3\varepsilon_0 (N_3/N)^2 J_1 J_2. \quad (46)$$

Thus, the asymptotic formulas for the bound-state energy and the magnon number  $N$  are expressible in terms of the constants  $J_1$  and  $J_2$ —certain integrals of the function  $\Psi(x)$  determined by Eq. (41). In this case it turns out that there exists between the constants  $J_1$  and  $J_2$  a relation that allows us, in computing  $E$  and  $N$ , to find by numerical methods only one of these constants.

To obtain the relation between  $J_1$  and  $J_2$ , it is sufficient to use the relation (27). Using the formulas (44) and (45), we obtain that  $J_1 = J_2$ . Taking this circum-

stance into account, we arrive at the conclusion that the localized state for  $\omega_0 - \omega \ll \omega_0$  possesses the following characteristics:

$$\begin{aligned} E/N &= \varepsilon_0 \{1 + (N_3 J_1 / N)^2\} > \varepsilon_0, \\ \omega &= \omega_0 \{1 - (N_3 J_1 / N)^2\} < \omega_0. \end{aligned} \quad (47)$$

The condition  $\omega_0 - \omega \ll \omega_0$  corresponds to the requirement that  $N \gg N_3$ . Thus, there exist two types of localized states corresponding to  $N \gg N_3$ : the "hard" state ( $\omega \ll \omega_0$ ) considered by us above, in which there exists the region  $\theta(r) \approx \pi$  and the magnetization varies in a narrow interval of variation of the variable  $r(\Delta r \sim l)$  and a "smeared-out" state:  $\omega_0 - \omega \ll \omega_0$  (see Fig. 4).

To elucidate the character of the self-localized solutions to Eq. (28) and analyze the dependence  $E(N)$  for arbitrary values of the parameter  $\omega$ , we carried out a numerical integration of this equation.<sup>6)</sup> The plots in Fig. 4 give an idea of the magnetization distribution in the bound state at high  $\omega$ . This same figure demonstrates the validity of our analysis of the self-localized solution for  $\omega \ll \omega_0$  and the spatially "smeared out" nature of the solution in the case when  $\omega_0 - \omega \ll \omega_0$ .

The results of the numerical computation of the dependences  $\omega = \omega(N)$  and  $E = E(N)$  are presented in Figs. 5 and 6. It turned out that  $\omega$  and  $E$  are two-valued functions of  $N$ . This indicates the existence of two bound-state branches. The physical meaning of this result amounts to the assertion that in the three-dimensional case  $N$  magnons can form bound states of two types that differ in, besides the precession frequency and the energy, the nature of the spatial distribution of the magnetization.

In the low-frequency states the region of inhomogeneity of the magnetization on the  $r$  axis does not exceed in order of magnitude the thickness of the static domain wall. The high-frequency oscillations have a feebly marked localization, and the characteristic dimension of the region of localization increases in inverse proportion to the quantity  $\sqrt{\omega_0 - \omega}$  as  $\omega \rightarrow \omega_0$ . As it turned out, the plot of the function  $\omega(N)$  has a vertical tangent at  $\omega = \omega_* = 0.915$ ,  $N = N_* = 9.08$ , and it is well approximated near this point by a quadratic parabola. Using the latter fact and the relation (27), we can easily show that there obtains near the end point of the plot of  $E(N)$ , i. e., near the point  $N = N_*$ , the expansion

$$E(N) = E(N_*) + \hbar\omega_0 (N - N_*) \pm A(N - N_*)^2, \quad (48)$$

in which  $E(N_*) = 1.034N_*\varepsilon_0$  and  $A = \text{const}$ . The plus and

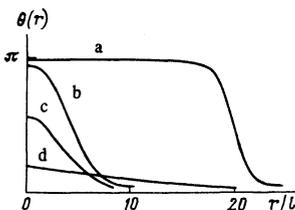


FIG. 4. Dependence of the precession angle  $\theta$  on  $r$  in a three-dimensional ferromagnetic substance: a)  $\omega = 0.1\omega_0$ , b)  $\omega = 0.5\omega_0$ , c)  $\omega = \omega_* = 0.915\omega_0$ , d)  $\omega = 0.99\omega_0$ ;  $l = \sqrt{\alpha/\beta}$ .

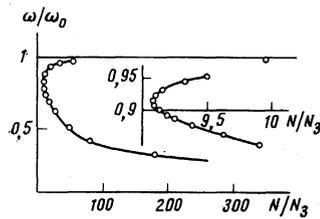


FIG. 5. Dependence of the precession frequency,  $\omega(N)$ , on the magnon number in the bound state. The points represent the results of the numerical calculation.

minus signs in (48) pertain to the upper and lower bound-state branches.

The  $N$ -magnon bound states pertaining to the lower branch are stable against small perturbations. It is clear that states for which decay into magnon states of the continuous spectrum is forbidden will be stable against arbitrary perturbations (let us recall that the Hamiltonian (14) commutes with the  $z$  component of the total spin and that the number of magnons is conserved). The states of the upper branch are, as follows from Ref. 15, unstable, while the states of the lower branch are metastable when  $N < N_0 = 11.3N_3$ . Notice that both stable and metastable  $N$ -magnon bound states are possible in a three-dimensional ferromagnet only when  $N \geq N_* \gg 1$ . The latter justifies the use of the quasi-classical approximation to describe the  $N$ -magnon bound states in a three-dimensional ferromagnet with a weak anisotropy (let us recall that the condition  $\mu_0\beta M_0 \ll I$  corresponds to  $N_3 \gg 1$ ).

It was shown in Ref. 7 that the bound states of two magnons with zero total quasimomentum are formed when the condition  $\mu_0\beta M_0 \geq \eta I$ , where  $\eta$  is a numerical parameter of the order of unity, is fulfilled. Under such a condition the characteristic number,  $N_3$ , introduced by us is of the order of unity, and, therefore, our results are not at variance with the results of Ref. 7.

In conclusion of the present section, let us note that we can easily take into account an external uniform magnetic field  $H$  directed along the easy axis. Then to the energy of the ferromagnet must be added the usual Zeeman energy of magnetization in an external field:

$$W_H = 2\mu_0 H N.$$

When written in such a way, the Zeeman energy is measured from the energy of the homogeneous magnetized state of the ferromagnet. To the positive value of  $H$  corresponds the parallel orientation of the magnetic field and the magnetization of the ground state of the ferromagnet. It is not difficult to verify that the equa-

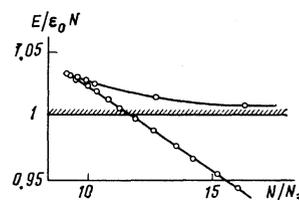


FIG. 6. Dependence of the energy per magnon in the bound state on the number,  $N$ , of magnons (the three-dimensional case).

tions for the magnetization distribution in the presence of a magnetic field differ from the Eqs. (19) and (28) only by the fact that the quantity  $\omega$  is replaced by the quantity  $\omega = \omega - \omega_0(H/H_A)$ , where  $H_A = \beta M_0$  is the field of the anisotropy. Taking this fact into account, we can easily picture the plots of the dependences  $\omega(N)$  and  $E(N)$  in the presence of an external magnetic field. The plot of the dependence  $\omega(N)$  for  $H \neq 0$  is obtained by simply shifting the plot in Fig. 5 upward by the amount  $\omega_0(H/H_A)$ , while the plot of the dependence  $E(N)/N$  for  $H \neq 0$  is obtained by shifting the plot in Fig. 6 upward by  $2\mu_0 H$ .

It is interesting that for  $H < 0$  there exist static equilibrium states of the ferromagnetic substance with a finite number of spin deviations. To elucidate the meaning of these states, it is necessary to note that there exist in an external field parallel to the anisotropy axis ( $H < H_A$ ) two homogeneous stable states of the ferromagnet: a stable state when  $H > 0$  and a metastable state when  $H < 0$ , i. e., in the case of antiparallel orientation of the magnetic field and the magnetization.

The static inhomogeneous solution with a finite number of spin deviations found by us to exist in the case when  $H < 0$  is, in fact, the nucleus of a region with opposite magnetization direction in an unbounded single-domain ferromagnet in a metastable state ( $H < 0$ ), i. e., the usual critical nucleus of the stable phase in a metastable phase.<sup>7)</sup>

Turning to the interpretation of the macroscopic meaning of the investigated self-localized states with  $\omega \neq 0$ , let us note the following. First, if the ensemble of "free" magnons can be treated as a gas of quasiparticles with a weak attractive interaction (the energy of the magnetic anisotropy of the easy-axis type corresponds to a two-body attractive interaction between long-wave magnons), then the bound state of a large number of magnons is a "magnon drop." It is clear that, when the relaxation processes are taken into account, such a state of the magnetic substance can exist only under the conditions of an external influence that guarantees the prescribed number,  $N$ , of spin deviations. Secondly, for the existence of the magnon drop, it is necessary that the mean lifetime of the magnons be longer than the time required for their condensation into a drop and that the resorption of the drop on account of the relaxation mechanisms in the magnetic substance be compensated by the excitation of magnons by the external influence.

## CONCLUSION

While analyzing the question of the existence of magnon bound states in a three-dimensional ferromagnetic substance, we showed that, even if two-magnon bound states cannot be formed in the ferromagnet, the formation of bound states of a large number of magnons is always possible. It seems to us that it is useful to formulate this result in a more general form.

The amplitude,  $U$ , of the interaction of the long-wave magnons is determined by the magnetic-anisotropy energy, and the condition  $\mu_0 \beta M_0 \ll I$  can be qualitatively

interpreted as the condition for the smallness of the magnon-interaction energy as compared to the energy band width,  $\delta \varepsilon \sim I$ , of the free magnon. It is well known that quasiparticle bound states are formed only in the case when the amplitude,  $U$ , of their attractive interaction exceeds some critical value  $U_c$ , where  $U_c \sim \delta \varepsilon$ . The inequality  $\mu_0 \beta M_0 \ll I$  for magnons in a ferromagnet of the easy-axis type implies that  $U \ll U_c$ , and, therefore, the absence of two-magnon bound states is not surprising.

However, as we have shown, there exist in a ferromagnet with a small anisotropy  $N$ -magnon bound states with  $N \geq N_* \sim (I/2\mu_0 \beta M_0)^{3/2}$ , which can be rewritten as  $N \geq N_* \sim (U_c/U)^{3/2}$ .

Therefore, we can apparently make the following general assertion concerning any Bose quasiparticles in a three-dimensional crystal: if the magnitude of the attractive-interaction potential,  $U$ , of the bosons is not sufficient for the formation of bound pairs ( $U < U_c$ ), then  $N$ -particle bound states can be formed when  $N > N_c \sim (U_c/U)^{3/2}$ .

The authors are profoundly grateful to K. V. Maslov for a discussion of a number of mathematical aspects of the problem and for his help in the formulation of the numerical computations, as well as to A. A. Motorna and V. I. Khatuntsev for carrying out the computer calculation.

*Note added in proof (March 28, 1977).* Recently, V. I. Petviashvili drew our attention to a paper by N. G. Vakhilov and A. A. Kolokolov (Izv. Vyssh. Uchebn. Zaved. Radiofiz. 16, 1020 (1973)), in which it is shown that a necessary condition for the stability of a localized solution of an equation of the type (41) is the fulfillment of the inequality  $d\omega/dN < 0$  (in our notation). On this basis we draw the conclusion that the states corresponding to the upper branches in Figs. 4 and 5 are unstable. We are grateful to V. I. Petviashvili for the very useful remark.

- <sup>1)</sup>It should be noted that the single-ion anisotropy energy (1) does not make any contribution to the amplitudes of the interaction of more than two magnons, and for  $k \rightarrow 0$  the dominant term in the Hamiltonian is the magnon-magnon interaction (4).
- <sup>2)</sup>The fact that the magnetic dipole interaction does not conserve the magnon number is of little consequence. As follows from the results of Ref. 9, the properties of the self-localized states and their classification should, in the first approximation in the small parameter  $4\pi/\beta$ , not change.
- <sup>3)</sup>We intentionally denoted the coordinate along the chain by the Greek letter  $\xi$ , and not by the letter  $x$ , as was done in the preceding section, so as not to create the impression that the anisotropy axis is necessarily perpendicular to the axis of the chain.
- <sup>4)</sup>The energy of the self-localized oscillation is reckoned from the ground-state energy of the ferromagnetic substance.
- <sup>5)</sup>If we take  $\tau$  to be the time, then Eq. (19) coincides with the equation of motion of a nonlinear conservative oscillator. In contrast to (19), Eq. (28) includes a frictional force proportional to the "velocity"  $\theta'$ , and the relation (30) determines the standard connection between the variation of the energy of the system and the dissipative function of the non-

conservative nonlinear oscillator.

- <sup>6</sup>The numerical computations were carried out at the computing center of the Physicotechnical Low-Temperature Institute of the Academy of Sciences of the Ukrainian SSR by A. A. Motornaya and V. I. Khatuntsev.
- <sup>7</sup>Such a magnetization distribution resembles a "spherical domain." We should only bear in mind that, in contrast to the conventional theory of cylindrical domains in thin films, our analysis has been carried out without allowance for the magnetic-dipole interaction.
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Translated by A. K. Agyei

## Effect of atoms of second coordination sphere on the Mössbauer spectra of $^{119}\text{Sn}$ in diamondlike crystals

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(Submitted December 29, 1976)  
*Zh. Eksp. Teor. Fiz.* **72**, 2016-2021 (May 1977)

We investigated the Mössbauer spectra of  $^{119}\text{Sn}$  in solid solution of diamondlike semiconductors  $(\text{Cu}_2\text{SnS}_3)_x-(3\text{ZnS})_{1-x}$  ( $x = 1/16$  to 1) and  $(\text{Cu}_2\text{SnS}_3)_x-(3\text{CdS})_{1-x}$  ( $x = 3/16$  to 1). At values  $x < 3/4$ , when the probability of the appearance of the tin atoms in the second coordination sphere of the Mössbauer atom is low, the spectra consist of one somewhat broadened line, the parameters of which are practically independent of  $x$ . At values  $x \geq 3/4$ , when a certain fraction of the Mössbauer atoms in the second coordination sphere acquire tin atoms, the line begins to broaden and is transformed into a doublet with further increase of  $x$ , while the isomeric shift increases. The results show that large displacements of the electron charges can be induced by the atoms of the second coordination sphere in valence bonds of the atoms situated at the sites of a diamondlike crystal. The observed maximal values of the quadrupole splitting connected with the induce displacement of the electron charges are equivalent to the value of the electric field intensity gradient produced at the nucleus by approximately 1/3 of the unbalanced  $sp^3$  electron. The largest displacements, which lead to a measurable isomeric shift and to a quadrupole splitting of the Mössbauer spectrum line is to be expected in those cases when the second coordination sphere contains atoms elements from far removed groups of the periodic system.

PACS numbers: 76.80.+y

### 1. INTRODUCTION

The influence exerted on the structure of the Mössbauer spectrum by atoms that are not directly connected with the Mössbauer atom was observed in tin-organic compounds.<sup>[1,2]</sup> It was observed in those cases when an atom having large electronegativity or a group of atoms having strongly polar properties is joined to a ligand connected with the Mössbauer atom, and it was attributed to a displacement of the electronic charges of the molecule towards the electronegative center (to the induction effect).

Highly suitable objects for the study of the redistributions of the electronic charges due to the appearance

of a great variety of atoms near the Mössbauer atom and at various distances from it, are a complex diamondlike semiconductors.<sup>[3,4]</sup> In contrast to tin-organic compounds, where the interpretation of the results is frequently made difficult by the lack of structural data, the crystal structures of many diamondlike semiconductors are well known; short-range order is characterized by tetragonal symmetry, while in the case of long-range order the crystal lattices are most frequently of the sphalerite, chalcopyrite, or wurtzite type. In these cases when new complex compounds are synthesized or their solid solutions are produced, the crystal structure is easily obtained by x-ray diffraction. An exceptionally favorable circumstance is the