### Phase transition in system of nuclear spins with indirect Suhl-Nakamura interaction

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It is shown that under certain conditions, viz., 1) in a strong magnetic field which compensates the intrinsic longitudinal field at the nucleus or 2) under the action of a transverse alternating magnetic field with a frequency equal to the Larmor spin precession frequency, a first-order phase transition to the magnetically ordered state takes place in the nuclear spin system of ferromagnets or antiferromagnets (in the second case, the ordering occurs in a rotating coordinate system). Long-range order in the lattice is due to the Suhl-Nakamura interaction which, under various conditions, constitutes different examples of the XY model with long-range exchange integrals. The magnetization and spectra of the nuclear spin waves in the new phase are calculated. The parameters for which the phenomenon can be observed are estimated. The feasibility of its experimental realization is discussed.

#### **1. INTRODUCTION**

The indirect Suhl-Nakamura interaction, <sup>1,2</sup> which describes the coupling of transverse components of nuclear spins by means of virtual magnon exchange, plays an important role in the theory of nuclear magnetic resonance (NMR) in magnetic dielectrics. NMR line-shape moments<sup>3-5</sup> as well as more subtle effects associated with correlations in the motion of nuclear spins at distances of the order of the interaction range (nuclear spin waves<sup>5-7</sup>) are calculated in a relatively simple fashion on the basis of these interactions. We show below that under certain conditions the Suhl-Nakamura interaction in nuclear spin sytems may result in still another interesting effect: phase transition to a magnetically ordered state.

Consider the Hamiltonian for a system of nuclear spins in a ferromagnet<sup>5</sup>:

$$\mathcal{H} = -\hbar (AS - \mu_n H) \sum_{j} I_{j^z} - \frac{\hbar}{2} \sum_{j,j'} U_{jj'} (I_j^x I_{j'}^x + I_j^y I_{j'}^y),$$
$$U_{jj'} = \frac{A^2 S}{N} \sum_{\mathbf{k}} \frac{1}{\omega_k} \exp[i\mathbf{k} (\mathbf{R}_j - \mathbf{R}_{j'})]. \tag{1}$$

A is the constant of the hyperfine interaction between the nuclear spin I and the electron-shell spin S; H is the external magnetic field;  $U_{ii'}$  is the amplitude of the indirect interaction;  $\omega_k = \mu_e H + \alpha k^2$  is the frequency of a magnon with wave vector  $\mathbf{k}$ ;  $\mathbf{R}_i$  are the spin coordinates; N is the number of unit cells in the crystal;  $\mu_e$  and  $\mu_n$  are the gyromagnetic ratios for electron and nuclear spins. It follows from (1) that for A > 0 there exists a compensation point  $H_c = AS/\mu_n$ where the internal longitudinal field at the nuclei (which usually determines their ground state) vanishes and the Suhl-Nakamura Hamiltonian, which is an example of an XY-model with a particular exchange integral, becomes dominant. At sufficiently low temperatures this indirect spin-spin interaction produces in the nuclear-spin system a cooperative effect, consisting of ferromagnetic ordering in the xy-plane. A similar, but somewhat more complicated situation takes place in antiferromagnetic nuclear systems.

The Hamiltonian (1) describes also a magnetic sublattice formed by electron spins of rare-earth ions interacting with an ordered spin sublattice of iron ions, as in, for example, gadolinium iron garnets.<sup>8</sup> Thus, the results obtained below can be applied to this case as well.

The problem can be reformulated in such a way that instead of a large compensating field, it may be sufficient to apply a transverse alternating magnetic field or small amplitude with a frequency equal to the frequency of Larmor precession of the nuclear spins. The phase transition to the ordered state of nuclear spins coupled by the Suhl-Nakamura interaction takes place then in a rotating coordinate system.<sup>1)</sup> Ordering of nuclear spins in a weakly anisotropic antiferromagnet, in which the Suhl-Nakamura interaction is strengthen by the interplanar exchange, is of greatest interest in this case.

It should be mentioned that the possibility of a transition to a new nuclear spin state in magnetic materials near the region of internal magnetic field compensation was first pointed out by Tsifrinovich and Ignatchenko<sup>11</sup>. In their approach the problem was effectively reduced to calculating equilibrium configurations of nuclear and electron spins at the same site. This technique, naturally, excludes the possibility of analysis of collective effects in the system. A microscopic approach based on the indirect Suhl-Nakamura interaction allows one to describe the complete picture of the transition of a system of nuclear spins into a new phase.

# 2. EQUILIBRIUM CONFIGURATION OF NUCLEAR SPINS IN A FERROMAGNET

Assume that all nuclear spins are in the xz plane and make an angle  $\psi$  with the z axis. Then

$$I_j^{x} = I_j^{\zeta} \sin \psi + I_j^{\eta} \cos \psi, \ I_j^{z} = I_j^{\zeta} \cos \psi - I_j^{\eta} \sin \psi, \tag{2}$$

where  $\zeta$  and  $\eta$  are the quantization axes. Substituting this expression in the Hamiltonian (1) one can easily calculate an effective field acting on an individual spin. We set the transverse component of the field equal to zero and from that derive the equation for  $\psi$ :

$$\mu_n H_n = (AS - \mu_n H) \sin \psi - \langle I \rangle U_{SN} \sin \psi \cos \psi = 0, \qquad (3)$$

where  $U_{SN} \equiv \sum_{j} U_{jj'}$  and  $\langle I_{j}^{\zeta} \rangle \equiv \langle I \rangle$  is the average spin polarization determined by the field  $H_{\zeta}$ . Equation (3) has the following solutions:

1)  $\sin \psi = 0$ , (4a)

2) 
$$\cos \psi = (AS - \mu_n H) / \langle I \rangle U_{sN}.$$
 (4b)

It is clear that when  $|AS - \mu_n H| > \langle I \rangle U_{SN}$  only the first solution is valid. In this case  $\psi = 0$  for  $H < H_c$  and  $\psi = \pi$  for  $H > H_c$ , while the polarization is

$$\langle I \rangle = IB_I (I\hbar | AS - \mu_n H | / k_B T), \qquad (5)$$

where

$$B_{I}(x) = \left(1 + \frac{1}{2I}\right) \operatorname{cth}\left[\left(1 + \frac{1}{2I}\right)x\right] - \frac{1}{2I} \operatorname{cth}\left(\frac{x}{2I}\right)$$

is the Brillouin function.

In the parameter range where both solutions (4a) and (4b) exist, the energy minimum is achieved for the second solution, and then

$$\langle I \rangle = IB_{I}(I \langle I \rangle \hbar U_{sN} / k_{B}T).$$
(6)

The phase transition temperature  $T_{SN}$  is determined from Eq. (6). It is approximately equal to

$$k_{B}T_{SN} \approx \hbar \frac{I(I+1)}{3} U_{SN} \approx \hbar \frac{I(I+1)A^{2}S}{3\omega_{0}}.$$
 (7)

Note that this formula with allowance for  $\omega_0 \approx \mu_e H_c$  coincides with a similar expression from Ref. 11. This is due to the fact that the exchange field  $H_E$  does not enter in the amplitude  $U_{SN}$  and therefore does not introduce the specific features of ferromagnetic ordering of electron spins. Nevertheless expression (7) describes the characteristic temperature of the cooperative effect in a system of nuclear spins with large interaction radius  $r_{SN} \sim a(H_E/H_c)^{1/2}$  (*a* is the linear dimension of the cell).<sup>5</sup>

Let us estimate  $T_{SN}$  for the typical ferromagnet EuO  $(H_E \approx 408 \text{ kOe})$  which contains 100% of either <sup>151</sup>Eu or <sup>153</sup>Eu(I = 5/2). For typical values of parameters  $AS/2\pi \approx 141$  MHz, S = 7/2,  $\mu_n^{151}/2\pi \approx 1.05$  MHz/kOe and  $\mu_n^{153}/2\pi \approx 0.465$  MHz/kOe (Ref. 12) we get  $T_{SN} \approx 2.2 \times 10^{-6}$  K for  $H_c \approx 134$  kOe (<sup>151</sup>Eu) and  $T_{SN} \approx 10^{-6}$  K for  $H_c \approx 303$  kOe (<sup>153</sup>Eu). For comparison, the characteristic temperature for <sup>151</sup>Eu nuclear ordering by dipole-dipole interaction in the same lattice  $(a \approx 5.15 \text{ Å})$  is equal to  $T_D \approx I(I+1)\hbar^3 \mu_n^2 / 3a^3 k_B \approx 0.75 \times 10^{-8}$  K (Ref. 9). It must be noted that an infralow temperature in a nuclear system can be achieved by means of adiabatic (i.e., fast in comparison with the nuclear spin-lattice relaxation time) switching-on of  $H_c$ . In this case nuclear spins can be considered isolated from the lattice, and their temperature decreases by comparison with the initial temperature by  $\mu_n H_c / U_{SN} \sim \mu_e / \mu_n \sim 10^3$  times.

We derive the spectrum of the nuclear spin waves from the Hamiltonian (1) by expansion of the spin deflections using the Holstein-Primakoff approach. The part quadratic in the Bose operators has the following form:

$$\mathcal{H}^{(2)} = \hbar \sum_{\mathbf{k}} \left[ \mathcal{A}_{\mathbf{k}} a_{\mathbf{k}}^{+} a_{\mathbf{k}}^{+} \frac{1}{2} \mathcal{B}_{\mathbf{k}} (a_{\mathbf{k}} a_{-\mathbf{k}}^{-} + \mathbf{H.a.}) \right],$$
  
$$\mathcal{A}_{\mathbf{k}} = \left( AS - \mu_{n} H \right) \cos \psi + A^{2} S \langle I \rangle \sin^{2} \psi / \omega_{0}$$
  
$$-A^{2} S \langle I \rangle (1 + \cos^{2} \psi) / 2 \omega_{\mathbf{k}},$$
  
$$\mathcal{B}_{\mathbf{k}} = A^{2} S \langle I \rangle \sin^{2} \psi / 2 \omega_{\mathbf{k}}.$$
 (8)

The procedure for diagonalization of this quadratic form is well known (see for example Ref. 13). In particular, the frequency of the normal oscillations is calculated from the formula

$$\Omega_k = (\mathscr{A}_k^2 - \mathscr{B}_k^2)^{\frac{1}{2}}.$$

Substituting the coefficients  $\mathscr{A}_k$  and  $\mathscr{B}_k$ , corresponding to the phase (4a), we obtain the known expression for the nuclear-spin-wave spectrum (see Ref. 5). The excitation spectrum in the new phase (4b) becomes

$$\Omega_{k} = \frac{A^{2}S}{\omega_{0}} \langle I \rangle \left(1 - \frac{\omega_{0}}{\omega_{k}}\right)^{\prime h} \left[1 - \left(\frac{AS - \mu_{n}H}{A^{2}S \langle I \rangle}\right)^{2} \frac{\omega_{0}^{3}}{\omega_{k}}\right]^{\prime h}.$$
(9)

It is clear that this is a gapless spectrum. In the compensation point  $\Omega_k \propto k$ , while when  $|\cos \psi| = 1$  one has  $\Omega_k \propto k^2$ . Note also that the upper point of the spectrum  $\Omega_k = A^2 S \langle I \rangle / \omega_0$  lies in the region of relatively low frequencies (~100 kHz for EuO).

## 3. NUCLEAR-SPIN STATE NEAR THE COMPENSATION FIELD IN AN ANTIFERROMAGNET

Let us consider a system of nuclear spins of a two-sublattice antiferromagnet in a magnetic field that almost cancels the internal field at the nucleus. We assume that  $H_c > 2H_E$  when the system of ordered electron spins of the sublattices  $\{\mathbf{S}_g\}$  and  $\{\mathbf{S}_f\}$  are in a spin-flip phase in which the soft branch of spin waves is a spin flip (sf) mode with spectrum  $\omega_{sf}(k) = \mu_e (H - 2H_E) + \alpha k^2$  (see for example Ref. 14). The effective Hamiltonian  $\mathcal{H}_{eff}$  for nuclear system can be derived from the antiferromagnetic Hamiltonian

$$\mathscr{H} = \sum_{g,f} J(\mathbf{r}_{gf}) \mathbf{S}_{g} \mathbf{S}_{f} - \hbar \sum_{j=g,f} [\mathbf{H}(\mu_{g} \mathbf{S}_{j} - \mu_{n} \mathbf{I}_{j}) + A \mathbf{S}_{j} \mathbf{I}_{j}],$$
(10)

by taking into account the indirect nuclear spin-spin interaction caused by exchange of virtual *sf* magnons only. As the result we have<sup>2)</sup>:

$$\mathcal{H}_{eff} = -\hbar (AS - \mu_n H) \sum_{j=g,f} I_j^z - \frac{\hbar}{2} \sum_{\substack{j=g,f; \\ j'=g,f}} U_{jj'} (I_j^x I_{j'}^x + I_j^y I_{j'}^y),$$
$$U_{jj'} = \varphi(j,j') \frac{A^2 S}{N} \sum_{\mathbf{k}} \frac{1}{\omega_{sf}(k)} \exp[i\mathbf{k} (\mathbf{R}_j - \mathbf{R}_{j'})],$$
(11)

where  $\varphi = 1$  if the indices j and j' refer to spins on the same sublattice and  $\varphi = -1$  in the opposite case. It is clear from (11) that the Suhl-Nakamura interaction is the Hamiltonian of a two-sublattice XY model with exchange integrals that contribute to ferromagnetic ordering of nuclear spins belonging to the same sublattice and to antiferromagnetic ordering of spins from different sublattices.

Let us assume that all nuclear spins make an angle  $\psi$  with the z axis in one sublattice and  $-\psi$  in the other. Then the equilibrium condition in the mean-field approximation is:

$$(AS - \mu_n H) \sin \psi - \langle I \rangle U_{sN}^{(sf)} \sin \psi \cos \psi = 0,$$
$$U_{sN}^{(sf)} = \sum_{g'} U_{gg'} - \sum_{f} (U_{gf} + U_{fg})/2.$$
(12)

This equation is fully analogous to Eq. (3) for nuclear spins in ferromagnets, therefore formulas (4)–(6) are valid also in this case upon substitution of  $U_{SN}^{(sf)}$  for  $U_{SN}$ . The phasetransition temperature is equal to

$$k_B T_{SN}^{(\bullet)} \approx \hbar 2 I (I+1) A^2 S / 3 \omega_{sf}(0).$$
<sup>(13)</sup>

For the antiferromagnet MnSeO<sub>4</sub> ( $T_N \approx 20$  K,  $H_E \sim 200$  kOe) with <sup>55</sup>Mn nuclei (I = 5/2,  $AS/2\pi \sim 600$  kHz, and  $H_c \sim 600$  kOe) we get  $T_{SN}^{(sf)} \sim 10^{-4}$  K.

The small-oscillation spectrum of a nuclear system in an antiferromagnet is determined by the following quadratic form:

$$\mathcal{H}^{(2)} = \hbar \sum_{\mathbf{k}} \left\{ \mathscr{A}_{\mathbf{k}} (a_{\mathbf{k}}^{+}a_{\mathbf{k}}^{+}b_{\mathbf{k}}^{+}b_{\mathbf{k}}) + \left[ \frac{1}{4}\mathscr{B}_{\mathbf{k}} (a_{\mathbf{k}}a_{-\mathbf{k}}^{-} + b_{\mathbf{k}}b_{-\mathbf{k}}^{-}-a_{-\mathbf{k}}b_{\mathbf{k}} \right] + \mathscr{B}_{\mathbf{k}}a_{\mathbf{k}}^{+}b_{\mathbf{k}}^{-} + \mathbf{H.a.} \right\},$$

$$\mathcal{A}_{\mathbf{k}} = (AS - \mu_{n}H)\cos\psi + 2A^{2}S\langle I\rangle\sin^{2}\psi/\omega_{sf}(0) - \mathscr{B}_{\mathbf{k}},$$

$$\mathcal{B}_{\mathbf{k}} = A^{2}S\langle I\rangle\sin^{2}\psi/\omega_{sf}(k),$$

$$\mathscr{B}_{\mathbf{k}} = A^{2}S\langle I\rangle(1 + \cos^{2}\psi)/2\omega_{sf}(k),$$
(14)

where  $a_k^+$ ,  $a_k$  and  $b_k^+$ ,  $b_k$  are the Bose operators of the spin deviations in the  $\{g\}$  and  $\{f\}$  sublattices, respectively.

After the canonical transformation

$$a_{\mathbf{k}} = (d_{\mathbf{k}} + c_{\mathbf{k}})/\sqrt{2}, \quad b_{\mathbf{k}} = (d_{\mathbf{k}} - c_{\mathbf{k}})/\sqrt{2}$$
 (15)

expression (14) separates into two independent parts

$$\mathcal{H}_{1}^{(2)} = \hbar \sum_{\mathbf{k}} \left[ \left( \mathcal{A}_{\mathbf{k}} - \mathcal{C}_{\mathbf{k}} \right) c_{\mathbf{k}}^{+} c_{\mathbf{k}} + \frac{1}{2} \mathcal{B}_{\mathbf{k}} \left( c_{\mathbf{k}} c_{-\mathbf{k}} + \mathbf{H.a.} \right) \right], \quad (16a)$$

$$\mathscr{H}_{2}^{(2)} = \hbar \sum_{\mathbf{k}} \left( \mathscr{A}_{\mathbf{k}} + \mathscr{C}_{\mathbf{k}} \right) d_{\mathbf{k}}^{+} d_{\mathbf{k}}, \tag{16b}$$

from which the nuclear spin-wave spectra are easily found.<sup>3)</sup> The explicit form of these spectra for the new phase is:

$$\Omega_{1k} = 2 \frac{A^2 S}{\omega_{sf}(0)} \langle I \rangle \left[ 1 - \frac{\omega_{sf}(0)}{\omega_{sf}(k)} \right]^{\frac{1}{2}} \times \left[ 1 - \left( \frac{AS - \mu_n H}{2A^2 S \langle I \rangle} \right)^2 \frac{\omega_{sf}^{-3}(0)}{\omega_{sf}(k)} \right]^{\frac{1}{2}},$$

$$\Omega_{2k} = 2 \frac{A^2 S}{\omega_{sf}(0)} \langle I \rangle.$$
(17)

Just as in the ferromagnetic case, the low-frequency branch is not activated and at the compensation point we have  $\Omega_{1k} \propto k$ .

## 4. PHASE TRANSITION IN A ROTATING COORDINATE SYSTEM

Let us consider nuclear spins in a ferromagnet interacting with an external magnetic field within a time much shorter than the spin-lattice relaxation time. The Hamiltonian is then determined by the expression (1) and by the interaction energy

$$\delta \mathscr{H} = 2\hbar\omega_1 \cos \omega t \sum_j I_j^x, \tag{18}$$

where  $2\omega_1 = \mu_n h\eta$ ,  $\eta \equiv AS/\mu_n H$  is the gain,<sup>5</sup> and *h* is the field amplitude. It is convenient to describe the nuclear spin system in a coordinate system that rotates around the magnetic field direction with a frequency  $\omega$ . The transformation to such a representation is accomplished through the unitary operator  $U = \exp(-i\omega t \Sigma_j I_j^z)$  (see Ref. 9). As a result the nuclear spin Hamiltonian in a rotating coordinate system does not depend explicitly on time and takes the form<sup>17</sup>:

$$\tilde{\mathscr{H}} = -\hbar (AS - \mu_n H - \omega) \sum_{j} I_j^{z} + \hbar \omega_j \sum_{j} I_j^{x} - \frac{\hbar}{2} \sum_{j,j'} U_{jj'} (I_j^{x} I_{j'}^{x} + I_{j'}^{y} I_{j''}^{y}).$$
(19)

It is clear that the longitudinal component vanishes when  $\omega = \omega_c \equiv AS - \mu_n H$ . The field  $\omega_1/\mu_n$  then aligns the nuclear spins along the x axis. The average polarization is determined by the equation

$$\langle I \rangle = IB_{I}(I\hbar | \omega_{i} - U_{SN} \langle I \rangle | /k_{B}T_{s}).$$
<sup>(20)</sup>

Here  $T_s$  is the spin temperature in the rotating coordinate system and depends on the initial conditions and on the manner in which  $\delta \mathcal{H}$  (18) is applied.

From (20), under the condition that  $\omega_1 \ll U_{SN} \langle I \rangle$ , which is equivalent to  $\mu_e h \ll A \langle I \rangle$ , we get an expression similar to (7) for the nuclear-spin ordering temperature. An estimate of this temperature for EuO in a field  $H \sim 10$  kOe gives  $T_{SN} \sim 10^{-5}$  K, which is an order of magnitude larger than the estimate of the phase transition temperature in a strong compensating field. An additional decrease of the nuclear spin temperature by  $\sim (\omega_c - \omega_i)/U_{SN}$  times compared with the lattice temperature can be accomplished by means of an adiabatic change of field frequency from initial value of  $\omega_i$  to  $\omega_c$ .

The feasibility of experimental observation of the phase transition in a system of nuclear spins in a rotating coordinate system in weakly anisotropic antiferromagnets (e.g., RbMnF<sub>3</sub>,CsMnF<sub>3</sub>) or antiferromagnets with an easy plane anisotropy (MnCO<sub>3</sub>, etc.) is probably of greatest interest. In moderate magnetic fields the Suhl-Nakamura interaction in these systems is enhanced  $\omega_E/\omega_{fk} \ge 1$  times compared with a ferromagnetic system.<sup>5,6</sup> Here  $\omega_E \equiv \mu_e H_E$  and

$$\omega_{fk} = \mu_e \left[ H \left( H + H_D \right) + H_{\Delta}^2 + (\alpha k)^2 \right]^{\frac{1}{2}}$$

is the quasiferromagnetic spin-wave frequency,  $H_D$  is the Dzyaloshinskiĭ field, and  $H_{\Delta}^2$  is the hyperfine-interaction parameter. If a constant magnetic field is directed along x in the crystal basal plane xz, then, accurate to  $\sim \theta^2$  and  $(\mu_n H / AS)^2$  (where  $\theta \approx (H + H_D)/2H_E$  is the electronic spin sublattice magnetization cant angle), the effective Hamiltonian of a nuclear system can be expressed as (see Ref. 18):

$$\mathcal{H}_{eff} = -\hbar AS \left( \sum_{g} I_{g}^{z} - \sum_{f} I_{f}^{z} \right)$$
$$- \frac{\hbar}{2} \sum_{j,j'=g,f} \mathcal{D}_{jj'} (I_{j}^{z} I_{j'}^{s} + I_{j}^{y} I_{j'}^{y}),$$
$$\mathcal{D}_{jj'} = \varphi(j,j') \sum_{\mathbf{k}} \frac{AS - \omega_{nk}}{2N \langle I \rangle} \exp[i\mathbf{k} (\mathbf{R}_{j} - \mathbf{R}_{j'})], \qquad (21)$$

where  $\varphi = 1$  for j and j' belonging to the same sublattice, and  $\varphi = -1$  otherwise;

$$\omega_{nh} = AS(1 - 2A \langle I \rangle \omega_E / \omega_{fh}^2)^{\frac{1}{h}}.$$

The effective energy of interaction of nuclear spins with an alternating field  $h \cos \omega t$  ( $\mathbf{h} \| \hat{\mathbf{z}}$ ) can be expressed as

$$\delta \mathscr{H} = 2\hbar\mu_{eff}h\cos\omega t \left(\sum_{s} I_{s}^{x} - \sum_{f} I_{f}^{x}\right), \quad \mu_{eff} = \mu_{e}\theta \frac{AS\omega_{E}}{\omega_{f0}^{2}}.$$
(22)

Transforming the system described by the Hamiltonian (21) and (22) to a rotating coordinate system by means of the unitary operator

$$U = \exp\left[-i\omega t \left(\sum_{\sigma} I_{\sigma}^{z} - \sum_{r} I_{r}^{z}\right)\right]$$

we get the following effective Hamiltonian:

$$\widetilde{\mathscr{H}}_{eff} = -\hbar \left( AS - \omega \right) \left( \sum_{g} I_{g}^{z} - \sum_{f} I_{f}^{z} \right) \\ + \hbar \mu_{eff} \hbar \left( \sum_{\alpha} I_{g}^{x} - \sum_{f} I_{f}^{x} \right) \\ \cdot \qquad - \frac{\hbar}{2} \sum_{j,j'} \mathcal{O}_{jj'} \left( I_{j}^{x} I_{j'}^{x} + I_{j}^{y} I_{j'}^{y} \right).$$
(23)

It is easily seen that when  $\omega = \omega_c \equiv AS$  and  $\mu_{\text{eff}} h \ll \tilde{U}_{SN} \langle I \rangle$ , where

$$\overline{U}_{sN} = \sum_{s'} \overline{U}_{ss'} - \sum_{j} (\overline{U}_{sj} + \overline{U}_{js})/2 \approx (AS - \omega_{n0})/\langle I \rangle,$$

the antiferromagnetic-ordering temperature of the nuclear spins is

$$k_{B}\tilde{T}_{SN} \approx \hbar \frac{I(I+1)}{3} \tilde{U}_{SN}.$$
 (24)

The nuclear-spin-wave spectra in the new phase are

$$\Omega_{1k} = \Omega_{2k} (1 - \omega_{f0} / \omega_{/k})^{\frac{1}{2}}, \quad \Omega_{2k} = AS - \omega_{n0}.$$
(25)

It should be noted that a phase transition of a nuclear system must be accompanied by a sharp drop in the electronic-magnon activation energy  $\omega_{f0}$ , in which the contribution  $H^2_{\Delta}$  proportional to the longitudinal magnetization of the nuclear spins vanishes. This results in a temperature hysteresis typical of a first order phase transition.<sup>4)</sup>

Let us estimate  $\overline{T}_{SN}$  for MnCO<sub>3</sub> ( $H_E = 320$  kOe,  $H_D = 4.4$  kOe,  $AS/2\pi = 640$  MHz, S = I = 5/2,  $\mu_e H_{\Delta}^2$   $= 2H_E A \langle I^z \rangle$ ). For H = 0.5 kOe we get  $\overline{T}_{SN} \approx 30$  mK. By comparison, the temperature of the reverse phase transition is about 100 mK. This temperature range can be achieved experimentally (see Ref. 19). The temperature of the nuclear system can be lowered (as in the ferromagnetic case) by pulses of duration  $\tau(T_2 \ll \tau \ll T_1$ , where  $T_2$  and  $T_1$  are the nuclear spin-spin and spin-lattice relaxation times) at a frequency scan from  $\omega_i$  to  $\omega_c$ . The rise time  $\Delta \tau$  of these pulses must be short enough ( $\Delta \tau \ll T_2$ ) to prevent heating of the nuclei.

#### 5. CONCLUSION

In the earlier studies (Ref. 3–7,15,16,18) only the states of nuclear spins determined by the longitudinal field were considered, while the Suhl-Nakamura interaction played only the secondary role of establishing the correlations in the spin oscillations. This situation is, to a certain extent, analogous to a paramagnetic system with exchange interaction, located in a strong magnetic field. Naturally, there are no temperature anomalies in such a system: as temperature is lowered, spin polarization gradually increases, the thermal fluctuations subside, and the spin-wave region expands from the long wavelength spectral region.

Conditions under which the behavior of nuclear spins is determined by the indirect Suhl-Nakamura interaction, leading to a phase transition into a magnetically ordered state, were investigated in the present work. The nuclear systems discussed are examples of a three-dimensional XYmodel with long range  $(r_{SN} \ge a)$  exchange interaction. The long range of the interaction allows the utilization of the mean-field approximation, with results accurate of up to  $\sim (a/r_{SN})^3$ . The nuclear spin-wave spectra in the new phase usually have frequencies much lower than the characteristic precession frequency of nuclear spins in a hyperfine field, and this allows study of these excitations in rf range. An important factor is the possibility of studying the discussed phase transition in a rotating coordinate systems by means of pulsed NMR methods.

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- <sup>3)</sup>The nuclear spin-wave spectrum in a phase with sin  $\psi = 0$  was calculated in Refs. 15 and 16.
- <sup>4)</sup>A similar effect should also be observed in cases considered above, but the hysteresis in them is small because of the small contribution of the hyperfine interaction to the electronic-magnon spectrum.
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<sup>&</sup>lt;sup>2)</sup>It is assumed that  $AS \ll \mu_e (H_c - 2H_E) \ll \mu_e H_c$ .