

Dynamic properties of the nuclear subsystem in antiferromagnets

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A theoretical investigation is made of the relaxation processes in the nuclear subsystem of an easy-plane antiferromagnet under conditions of an arbitrary dynamic frequency shift in the spectrum of nuclear spin waves. The diagram technique for the spin operators is used to calculate the relaxation frequencies of nuclear spin waves both in the range $T \gg \omega_n$ and in the range $T \ll \omega_n$ (ω_n is the undisplaced nuclear magnetic resonance frequency). It is shown that in the case of a large dynamic frequency shift it is necessary to allow not only for the pair (Suhl-Nakamura) interaction of nuclear spins with one another, but also for many-spin interactions. When the latter interactions are allowed for in a suitable manner in the case of basal plane symmetry, the spectrum of nuclear spin waves exhibits a hydrodynamic region ($\omega_k \propto k$) where the damping decrement of nuclear spin waves is governed by four-wave scattering processes: $\gamma \propto k^2$. At temperatures $T \gtrsim \omega_n$ the main contribution to the damping of nuclear spin waves in the principal part of the phase space of the nuclear subsystem is made by the process of the scattering of these waves by thermal fluctuations of the longitudinal component of the nuclear spins: $\gamma_{fl} \propto kT\omega_k^3$. A determination is made of a characteristic temperature $T^* \gg \omega_n$ at which perturbation theory ceases to be valid in respect of the number of loops in the diagram and the spin-wave picture of the nuclear subsystem no longer applies. An allowance is made for the influence of the dipole-dipole interaction on fluctuation damping of nuclear spin waves.

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1. INTRODUCTION

Our aim will be to consider the relaxation processes involving nuclear spin waves exhibiting strong dispersion in antiferromagnets with the easy-plane anisotropy. In view of the strong exchange enhancement of the hyperfine interaction, the dynamic frequency shift in the spectrum of nuclear spin waves in substances of this kind may be of the order of the undisplaced NMR frequency ω_n . The large shift affects the dynamics of the nuclear subsystems as follows:

- 1) the nuclear spin-spin interactions of higher order than the Suhl-Nakamura (pair) interaction become important¹;
- 2) at temperatures $T \ll \omega_n$ the dominant contribution to the relaxation frequencies of nuclear spin waves are made by the processes of the scattering of these waves by one another.

We shall begin with the Hamiltonian of an easy-plane antiferromagnet

$$\hat{H} = \hat{H}_m + \hat{H}_n + \hat{H}_{m-n}, \quad (1.1)$$

where

$$\begin{aligned} \hat{H}_m = & -H \left(\sum_f S_f^x + \sum_g S_g^x \right) \\ & + \sum_{g,f} \{ J_{gf} S_g S_f + \beta_{gf} S_g^y S_f^y + d_{gf} [S_g, S_f]_y \} \end{aligned} \quad (1.2)$$

describes the subsystem of electron spins in two sublattices f and g , coupled by the exchange interaction with the anisotropy $\beta > 0$, and

$$\begin{aligned} \hat{H}_n + \hat{H}_{m-n} = & -\mu_n H \left(\sum_f I_f^x + \sum_g I_g^x \right) \\ & + A \left(\sum_f S_f I_f + \sum_g S_g I_g \right) \end{aligned} \quad (1.3)$$

describes the subsystem of nuclear spins subject to an allowance for the hyperfine interaction with the electron subsystem. In the low-temperature range $T \ll T_N$ the electron spins are ordered in the basal plane (x, z) at a small angle to the z axis, given by $\theta \approx (H + H_D)/2\omega_E$, where $H_D = Sd_0$ is the Dzyaloshinskii field and $\omega_E = SJ_0$ is the exchange frequency. Here and later we shall use the energy system of units: $k_B = \hbar = g\mu_B = 1$. In view of the smallness of the nuclear magneton μ_n we shall ignore the direct interaction of the nuclear spins with a magnetic field.

It is known that the spectrum of the electron magnons in an easy-plane antiferromagnet has two branches,^{2,3} one of which (corresponding to oscillations of the magnetization in the basal plane) is characterized by a low activation energy:

$$\left. \begin{aligned} \epsilon_h &= [\epsilon_0^2 + (sk)^2]^{1/2}, & \epsilon_0^2 &= \tilde{\epsilon}_0^2 + 2bJ_0\omega_n, \\ \tilde{\epsilon}_0^2 &= H(H+H_D)+\Delta^2. \end{aligned} \right\} \quad (1.4)$$

The formula (1.4) allows for the static influence of the nuclear spins with the polarization $b = \langle I^z \rangle_0 = IB_I(I\omega_n/T)$, where $\omega_n = AS$ is the undisplaced NMR frequency and $B_I(x)$ is the Brillouin function, and also for the contribution made to the energy gap of magnons by other effects (magnetoelastic interaction, anisotropy in the basal plane, etc.), denoted by Δ in the above equations.¹⁾

The spectrum of excitations of the nuclear subsystem of an easy-plane antiferromagnet also consists of two branches,^{4,5} one of which (lower) is associated with low-activation-energy magnons and exhibits strong dispersion:

$$\omega_h = \omega_n [1 - (2bJ_0\omega_n/\epsilon_h^2)]^{1/2}. \quad (1.5)$$

If the symmetry in the basal plane is not disturbed ($\tilde{\epsilon} = 0$), this branch or mode is of the Goldstone type with a linear dispersion law in the limit $k \rightarrow 0$.

At sufficiently high temperatures (or in sufficiently strong magnetic fields) when the dynamic frequency shift can be ignored, the experimental data on the NMR line width are described satisfactorily by the second Suhl-Nakamura moment¹

$$\Gamma_{SN} = (\Delta\omega_{SN})^{1/2}, \quad \Delta\omega_{SN}^2 = \frac{b'\eta^3}{16\pi S^2} \omega_n^2 \frac{\omega_n^2}{\varepsilon_0\omega_E}. \quad (1.6)$$

Here, $b' \approx I(I+1)/3$ is the derivative of the Brillouin function and η is the structure constant of the lattice ($\nu = 2^{1/3} \cdot 3^{1/2}$ for a simple cubic lattice and $\eta = 2$ for a bcc lattice). In the range $T \lesssim T^*$, where

$$T^* \approx [I(I+1)]^{1/2} \omega_n (\omega_E/\varepsilon_0)^{1/2}, \quad (1.7)$$

the dynamic frequency shift governed by the quantity $b \approx b'\omega_n/T$ becomes greater than Γ_{SN} and this gives rise to wave properties in the nuclear subsystem and to a considerable reduction in the damping. The relevant result for the damping decrement of nuclear spin waves γ_k , first obtained using the relaxation function method,⁶ is

$$\gamma_k = \frac{\eta^3}{8\pi S} \omega_n \frac{Tsk}{\omega_E^2} \quad (1.8)$$

and in the range $\omega_n - \omega_k \ll \omega_n, \omega_n \ll T \ll T^*$ it is confirmed by the experimental data on the parametric excitation of nuclear spin waves.⁷ As shown in Ref. 8, Eq. (1.8) describes damping of nuclear spin waves associated with their scattering by thermal fluctuations of the longitudinal component of the nuclear spins.

An important factor which makes it easier to study theoretically the properties of the nuclear subsystem is the fact that, because of its low energy ($\omega_n = 3 \cdot 10^{-3} - 3 \cdot 10^{-2} K \ll \varepsilon_0$) the subsystem does not have a significant dynamic effect on the electron subsystem. If we know the Green functions of magnons and the corresponding vertices, we can derive the effective Hamiltonian describing the nuclear subsystem in terms of the indirect interactions of the nuclear spins with one another via virtual magnons. It is then found that in the limit of a small dynamic frequency shift the dynamic Hamiltonian need contain only the indirect pair interaction (Suhl-Nakamura interaction) which gives rise to the spectrum of nuclear spin waves given by Eq. (1.5) and also yields the damping results given by Eq. (1.6) and (1.8). The situation becomes more interesting in the case of a large dynamic frequency shift when the amplitudes of the scattering of nuclear spin waves include a considerable contribution from the indirect interactions of higher orders. In the Goldstone limit ($\omega_0 \rightarrow 0$) an allowance for the latter results in significant contraction in the relevant amplitudes.^{8,9} However, a consistent analysis of these topics has not yet been carried out.

A rigorous approach to the dynamic properties of the nuclear subsystem requires application of the diagram technique for the spin operators²⁾ described in Refs. 10–12. This approach was used in Refs. 9 and 13 to calculate the lifetime of nuclear spin waves associated with the processes of their scattering by one another in the case of a small dynamic frequency shift for vectors obeying $sk \ll \varepsilon_0$. However, in the case of a large dynamic frequency shift the application of the diagram spin technique has until recently been hindered by

the absence (within the framework of this technique) of a diagonalization procedure similar to the canonical Bogolyubov transformation for the Hamiltonians of Fermi and Bose particles. The absence of a diagonalization procedure has made it necessary to use a cumbersome and ineffective matrix method.

We shall consider theoretically the dynamic properties of the nuclear subsystem of an easy-plane antiferromagnet with an arbitrary dynamic frequency shift using the procedure for the diagonalization of the spin Hamiltonian suggested in Ref. 14. We shall calculate the temperature renormalization of the spectrum and the relaxation frequencies of nuclear spin waves in a wide range of wave vectors both in the case when $T \gg \omega_n$ and also when $T \ll \omega_n$. When $T \ll \omega_n$, thermal fluctuations are weak and the dominant role is played by the processes of the scattering of nuclear spin waves by one another. If $T \gtrsim \omega_n$, then in the principal part of the phase space the main contribution to the damping of nuclear spin waves is made by fluctuation processes [see Eq. (3.2)]. In the case of a small dynamic frequency shift this part of the phase space coincides with the region of existence of nuclear spin waves found from the condition $\gamma_k \leq k \partial \omega_k / \partial k$ and having the form $k_1^* \leq k \leq k_2^*$, where

$$\left. \begin{aligned} k_1^* &\approx k_0 (T/T^*)^2, \\ k_2^* &\approx k_0 (T^*/T)^{1/2} = [I(I+1)]^{1/2} (\omega_n/T)^{1/2} k_{max} \end{aligned} \right\} \quad (1.9)$$

and $sk_0 = \varepsilon_0$ and $sk_{max} = \omega_E$. At $k \sim K_{1,2}^*$ the contributions of the processes of different orders (fluctuation, four-wave, six-wave, etc.) to the damping become comparable and perturbation theory in respect of the number of loops in the diagrams (expansion in terms of the excitation density) ceases to be valid. In the range $k \leq k_1^*$ and $k \geq k_2^*$ a situation typical of a paramagnet is encountered. Well inside the spin-wave range ($k \sim k_0$) a perturbation theory series converges in respect of the parameter $(T/T^*)^2 = (r_2^*/r_0)^3$, where $r_0 = 1/k_0$ is the radius of the Suhl-Nakamura interaction and $r_2^* = 1/k_2^*$.³⁰ When temperature is increased to $T \sim T^*$, the boundaries come closer together: we then have $k_1^* \sim k_2^* \sim k_0$ and the spin-wave part of the phase space of the nuclear subsystem disappears.

We shall also consider the influence of the dipole-dipole interaction between the electron spins on the spectrum and damping of nuclear spin waves. We shall show that corrections to the spectrum of nuclear spin waves due to the dipole-dipole interaction and resulting in the anisotropy of the spectrum in the \mathbf{k} space are proportional to the skew angle of the electron spin sublattices, i.e., to the degree of decompensation of the magnetic fields created by these sublattices. In the long-wavelength part of the phase space of the nuclear subsystem a characteristic dipole-dipole region with $k \leq k_d$ is observed⁴⁾ and in this region we have

$$(sk_d)^2 = \frac{4\pi}{\delta} (H + H_D)^2, \quad \delta = \frac{\omega_E}{M_0}, \quad (1.10)$$

where M_0 is the magnetization of the electron sublattice in which the dispersion of the nuclear spin wave spectrum $k |\partial \omega_k / \partial k|$ is governed entirely by the dipole-dipole interaction and does not disappear even in the limit $k \rightarrow 0$. The last circumstance stabilizes the spin-wave pattern which exists if

an allowance is made for the dipole-dipole interaction throughout the range $0 < k \leq k_2^*$ right up to the temperature

$$T_d^* \approx T \left[\frac{4\pi}{\delta} \left(1 + \frac{H_D}{H} \right) \right]^{1/4}, \quad (1.11)$$

at which $k_1^* \sim k_d$. When T_d^* is attained, the spin-wave picture no longer applies anywhere in the dipole-dipole region. One should point out that if an allowance is made for the dipole-dipole interaction in the most thoroughly investigated temperature range $\omega_n \lesssim T \lesssim T_d^*$, the fluctuation mechanism of the damping of nuclear spin waves plays the dominant role throughout the phase space $0 < k \leq k_2^*$ [see Eqs. (4.5)-(4.7)].

The present paper is organized as follows. In Sec. 2 we shall give the effective Hamiltonian of the nuclear subsystem of an antiferromagnet. We shall describe the diagonalization procedure which makes it possible to obtain a nuclear spin wave spectrum and the amplitudes of the interactions of spin waves in the case of an arbitrary dynamic frequency shift. In Sec. 3, we shall calculate the lifetimes of nuclear spin waves and renormalize the spectrum of these waves. In Sec. 4, an allowance is made of the influence of the dipole-dipole interaction on the spectrum and fluctuation damping of nuclear spin waves. The small parameters of the theory are refined in Sec. 5 and an estimate is obtained of the contributions made to the dynamics of the nuclear subsystem by processes that have not been allowed for so far. The results obtained are compared with the experimental data in the Conclusions.

2. HAMILTONIAN OF THE NUCLEAR SUBSYSTEM

The effective Hamiltonian of the nuclear subsystem of an antiferromagnet found by eliminating the electron degrees of freedom from Eq. (1.1) and containing variables describing only the lower branch of nuclear spin waves is as follows in the Fourier representation:

$$\begin{aligned} \hat{H} = & -\omega_n m_0^z - \frac{1}{N} \sum_{12} \delta(1+2) V_1 m_1^x m_2^x \\ & - \frac{1}{N^2} \sum_{123} \delta(1+2+3) U_{12} m_1^x m_2^x \delta m_3^z \\ & - \frac{1}{N^3} \sum_{1234} \delta(1+2+3+4) W_{1234} m_1^x m_2^x m_3^x m_4^x, \end{aligned} \quad (2.1)$$

where

$$\begin{aligned} m^a &= I_f^a + I_g^a, \quad m_i^x = \frac{1}{2}(m_{k_i}^+ + m_{-k_i}^-), \\ \delta m_i^z &= m_{k_i}^z - 2bN\delta(k_i). \end{aligned}$$

The amplitudes of the indirect interaction of the nuclear spins are shown schematically in Fig. 1 and are given by the following expressions:

$$V_1 = \omega_n^2 J_0 / 2\epsilon_1^2 \quad (2.2)$$

is the amplitude of the Suhl-Nakamura interaction (Fig. 1a);

$$U_{12} = -\omega_n^2 J_0^2 / 2\epsilon_1^2 \epsilon_2^2 \quad (2.3)$$

is the amplitude of the three-spin interaction due to the hyperfine interaction of the longitudinal components of the electron and nuclear spins (Fig. 1b);

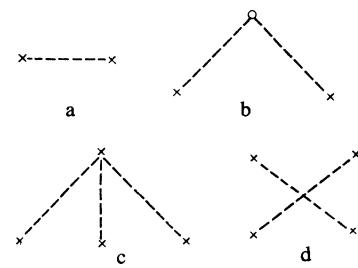


FIG. 1. Amplitudes of the indirect interaction between nuclear spins. The dashed lines represent the Green functions of the electron magnons, the circles show the longitudinal component of the nuclear spins, and the crosses give the transverse component of the nuclear spins.

$$W_{1234} = -\frac{\omega_n^4 J_0^3}{8\epsilon_1^2 \epsilon_2^2 \epsilon_3^2 \epsilon_4^2} \hat{S}_{1234} \left\{ \epsilon_1^2 + \frac{1}{3} (sk_1)^2 - H^2 - \tilde{\Delta}^2 \right\} \quad (2.4)$$

is the amplitude of the four-spin interaction due to the hyperfine interaction of the transverse components of the electron and nuclear spins (Fig. 1c) and also due to the four-wave anharmonicity of the electron subsystem (Fig. 1d). In Eq. (2.4) the quantity \hat{S}_{1234} is the operator for the symmetrization in respect of the wave vectors and the quantity $\tilde{\Delta}^2$ denotes the contribution made to the amplitude of the four-spin interaction by the elastic subsystem and other sources (see the Introduction), which will be specified later. In deriving the Hamiltonian (2.1) we have obtained only the terms up to the fourth order of magnitude in respect of the spin operators. On the other hand, in principle, the Hamiltonian contains terms also of higher orders, for example, of the sixth order in respect of the transverse components of the nuclear spins:

$$\begin{aligned} \hat{H}_6 = & -\frac{1}{N^5} \sum_{123456} \delta(1+2+3+4+5+6) \\ & \times W_{123456} m_1^+ m_2^+ m_3^+ m_4^+ m_5^- m_6^-, \end{aligned}$$

where

$$W_{123456} \sim \frac{\omega_n^6 J_0^5}{\epsilon_1^2 \epsilon_2^2 \epsilon_3^2 \epsilon_4^2 \epsilon_5^2 \epsilon_6^2} \hat{S}_{123456} \epsilon_1^2$$

[compare with the formula (2.4)]. Terms of this type make a contribution to the sixth-magnon processes which we shall not consider here. The contribution of these processes to the relaxation frequencies of nuclear spin waves should be small in respect of the parameter $(T/T^*)^2$ (see Sec. 5). Renormalizations of the four-magnon amplitudes due to such terms are represented graphically in Fig. 2 and they are small in respect of the parameter $(\omega_n/\epsilon_0)^2$. It should be pointed out that this is the adiabatic parameter that makes it possible to con-

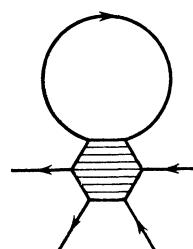


FIG. 2. Renormalization of a four-magnon amplitude.



FIG. 4. Amplitude of the scattering of nuclear spin waves by fluctuations of the longitudinal component of the nuclear spins. A semioval is used for the longitudinal correlation function of the nuclear spins. The last graph corresponds to the three-spin interaction.

which is quite cumbersome in the case of an arbitrary dynamic frequency shift (see the Appendix). We shall now give three limiting forms of this amplitude which will be used later.

1. Case of a small dynamic frequency shift ($\delta\omega_i = \omega_n - \omega_i \ll \omega_n$):

$$\begin{aligned}\Phi_{4n} &\approx -\frac{1}{4b}(\delta\omega_1 + \delta\omega_2 + \delta\omega_3 + \delta\omega_4) \\ &\approx -\frac{1}{4} \omega_n^2 J_0 \left(\frac{1}{\varepsilon_1^2} + \frac{1}{\varepsilon_2^2} + \frac{1}{\varepsilon_3^2} + \frac{1}{\varepsilon_4^2} \right).\end{aligned}\quad (2.17)$$

2. Case of a large dynamic frequency shift ($\omega_i \ll \omega_n$):

$$\Phi_{4n} \approx \frac{1}{8b\omega_n(\omega_1\omega_2\omega_3\omega_4)^{1/2}} \{2\tilde{\omega}_0^2\omega_n^2 - 3\omega_1\omega_2\omega_3\omega_4\}. \quad (2.18)$$

3. Case when two nuclear spin waves correspond to the range of small dynamic frequency shifts ($\delta\omega_{2,4} \ll \omega_n$) and two others to the range of large dynamic frequency shifts ($\omega_{1,3} \ll \omega_n$):

$$\Phi_{4n} \approx -(\omega_1\omega_3)^{1/2}/4b. \quad (2.19)$$

In Eq. (2.18) the quantity $\tilde{\omega}_0$ is given by the following expression when the magnetoelastic interaction is always allowed for:

$$\tilde{\omega}_0^2 = \left[H^2 + \frac{1}{4} HH_D + \Delta_{me}^2 \right] \frac{\omega_n}{2bJ_0}.$$

If $H_D = \omega_{mes} = 0$, the value of $\tilde{\omega}_0$ reduces to the energy gap in the spectrum of nuclear magnons ω_0 . It should be noted that in the case of a small dynamic frequency shift the scattering amplitudes Φ_A and Φ_{4n} are identical with those obtained allowing only for the Suhl-Nakamura interaction. On the other hand, in the case of a large dynamic frequency shift we have to allow for all the terms in the Hamiltonian (2.1). If the three- and four-spin terms are ignored, the Adler principle for the scattering amplitudes is violated and this is manifested in particular by a strong damping of the Goldstone excitations. The scattering amplitudes obtained in the present section will be used later for renormalization of the spectrum and in calculation of the relaxation frequencies of nu-

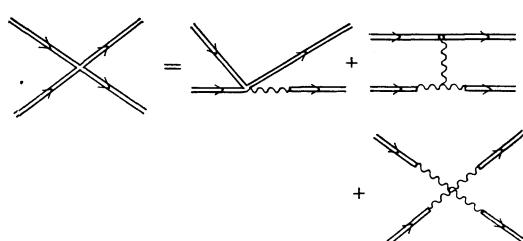


FIG. 5. Amplitude of the four-wave scattering of nuclear spin waves. The last graph corresponds to the four-spin interaction.

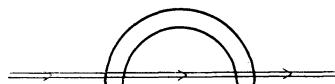


FIG. 6. Mass operator corresponding to the fluctuation scattering of nuclear spin waves. An oval is used for the longitudinal correlation function of the nuclear spins.

lear spin waves in a wide range of temperatures and wave vectors.

3. RELAXATION PROCESSES IN THE NUCLEAR SUBSYSTEM

The simplest of the processes resulting in relaxation in the nuclear subsystem is the fluctuation scattering of nuclear spin waves. The damping decrement of nuclear spin waves representing this process is governed by the imaginary part of the diagram shown in Fig. 6 and it is given by the expression

$$\gamma_{fl}(k) = 2b'v_0 \int \frac{d\mathbf{q}}{(2\pi)^3} |\Phi_{fl}|^2 \pi \delta(\omega_q - \omega_k), \quad (3.1)$$

where v_0 is the volume of a unit cell. A simple calculation gives the result

$$\gamma_{fl} = \frac{\eta^3}{8\pi S} \frac{b'}{b} \frac{\omega_k^3}{\omega_n} \frac{sk}{\omega_E^2}, \quad (3.2)$$

which generalizes Eq. (1.8) to the case of an arbitrary dynamic frequency shift. The presence in Eq. (3.2) of an additional [compared with Eq. (1.8)] factor $(\omega_k/\omega_n)^3$, which—in the Goldstone case—gives the dependence $\gamma_R \propto k^4$ eliminates the conflict with the results of a hydrodynamic theory of Ref. 16, according to which the damping of the Goldstone excitations with a linear dispersion law has the form $\gamma \propto k^2$ in the long-wavelength limit. We shall show later that the damping of the Goldstone nuclear spin waves is governed by the processes of their scattering on one another and is in agreement with the conclusions of the hydrodynamic theory.

The damping decrement of nuclear spin waves representing the four-wave scattering processes is governed by the imaginary part of the diagram expression given in Fig. 7. The diagram with a “paramagnetic” loop shown on the right of Fig. 7 making a negative contribution to the damping is related physically to the finite nature of the spin operator spectrum. Formally, the appearance of such diagrams is due to a supplementary procedure carried out in the course of construction of the Green spin-wave function in loops of spin-wave diagrams. The damping decrement of nuclear spin waves is

$$\gamma_{4n} = \gamma + \gamma^*, \quad (3.3)$$

where

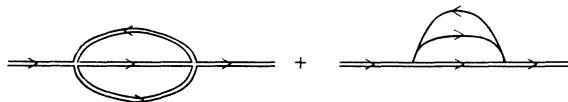


FIG. 7. Mass operator corresponding to the four-wave scattering of nuclear spin waves.

$$\gamma = \frac{1}{2n_k} v_0^2 \iint \frac{d\mathbf{p} d\mathbf{q}}{(2\pi)^3} |\Phi_{kn}(\mathbf{k}\mathbf{k}', \mathbf{pq})|^2 n_p n_q (n_k + 1) \pi \delta_{(\omega_p + \omega_q - \omega_k - \omega_h)} \quad (3.4)$$

$$\mathbf{k}' = \mathbf{p} + \mathbf{q} - \mathbf{k}$$

and

$$\gamma^* = -n_y (n_y + 1) v_0 \int \frac{d\mathbf{q}}{(2\pi)^3} |\Phi_{fl}|^2 \pi \delta(\omega_q - \omega_h). \quad (3.5)$$

The quantities $n_k = n(\omega_k/T)$, $n_y = n(\omega_n/T)$, etc., are the Bose occupation numbers related by $n(x) = (e^x - 1)^{-1}$. In Eq. (3.5) the variable in the integral is \mathbf{p} . We can see that the quantity γ^* can be expressed in a simple manner in terms of the fluctuation damping γ_{fl} [see Eq. (3.2)].

At low temperatures defined by $T \ll \omega_n$, where the quantities γ_{fl} and γ^* are exponentially small, the dominant contribution to the damping of nuclear spin waves is represented by Eq. (3.4). The interesting case is here that of a large dynamic frequency shift, when we can use a spectrum of long-wavelength nuclear spin waves of the type

$$\begin{aligned} \omega_k &= [\omega_0^2 + v^2 k^2]^{1/2}, \quad \omega_0^2 = \xi_0^2 \frac{\omega_n}{2bJ_0}, \\ v^2 &= s^2 \frac{\omega_n}{2bJ_0} \end{aligned} \quad (3.6)$$

and the scattering amplitudes (2.18) and (2.19). For the sake of brevity, we shall only give the results of calculations for $T \gg \omega_0$ [in the opposite limiting case the damping is exponentially weak: $\gamma \propto \exp(-\omega_0/T)$]. The relaxation frequencies of long-wavelength nuclear spin waves may be calculated using Eqs. (2.18) and (3.6). If $T \gg \omega_k$, the result is

$$\begin{aligned} \gamma_k &= \frac{b\eta^6}{3 \cdot 2^6 \pi^3 S^3} \frac{\omega_k \tilde{\omega}_0^4}{\omega_n \omega_E^3} \left(\frac{T}{\omega_k} \right)^2 \left\{ \Lambda \left(\frac{\omega_k}{\omega_0} \right) - 3^2 \cdot 2^6 C_1 \frac{T^2 \omega_k^2}{\tilde{\omega}_0^2 \omega_n^2} \right. \\ &\quad \left. + 3^3 \cdot 2^6 C_2 \frac{T^5 \omega_k^3}{\tilde{\omega}_0^4 \omega_n^4} \right\}, \end{aligned} \quad (3.7)$$

where $\Lambda(1) = 1$, $\Lambda(\infty) = 4$, and

$$\left. \begin{aligned} C_1 &= \iint_0^\infty dx dy \frac{xy(x+y)}{\sinh x \sinh y \sinh(x+y)} \approx 2.3, \\ C_2 &= \iint_0^\infty dx dy \frac{x^2 y^2 (x+y)^2}{\sinh x \sinh y \sinh(x+y)} \approx 6.4. \end{aligned} \right\} \quad (3.8)$$

The appearance of a negative term in the expression (3.7) is due to interference in the amplitude (2.18). We can see from Eq. (3.7) that the damping of a Goldstone nuclear spin wave ($\omega_0 \rightarrow 0$) is proportional to $\gamma \propto T^7 k^2$, in agreement with the hydrodynamic theoretical prediction of $\gamma \propto k^2$. In the opposite limiting case $T \ll \omega_k$, the damping decrement of nuclear spin waves is given by

$$\gamma_k = \frac{\pi b \eta^6}{400 S^3} \frac{T^4}{\omega_E^3} \left(\frac{\omega_k}{\omega_n} \right)^5 \left\{ 1 - \frac{50\zeta(3)}{\pi^4} \frac{\tilde{\omega}_0^2 \omega_n^2}{T \omega_k^3} + \frac{25}{12\pi^4} \frac{\tilde{\omega}_0^4 \omega_n^4}{T^2 \omega_k^6} \right\}. \quad (3.9)$$

The damping of short-wavelength nuclear magnons [characterized by $(sk)^2 \gg 2bJ_0 \omega_n$] with energies $\omega_k \approx \omega_n$ is due to their scattering by long-wavelength nuclear spin waves, which is of quasielastic nature.⁷⁾ A calculation carried out using the scattering amplitude (2.19) gives the result

$$\gamma_k = \frac{2\pi^3 b \eta^6}{24 S^3} \frac{T^7}{\omega_n^3 \omega_E^3}. \quad (3.10)$$

We shall now turn to high temperatures defined by $T \gg \omega_n$. In this case it is essential to include a "paramagnetic" diagram of Fig. 7. Its role reduces, roughly speaking, to truncation in the process of integration in Eq. (3.4) of a region in the phase space characterized by a small dynamic frequency shift, which has a significant influence on the damping results.⁸⁾ We shall first consider the relaxation of long-wavelength nuclear spin waves with a large dynamic frequency shift ($\omega_k \ll \omega_n$). In this situation it is not possible to calculate the damping decrement analytically, since excitations with an arbitrary dynamic frequency shift participate in the process and for these excitations the spectrum and the scattering amplitudes are quite complex [see Eq. (1.5) and the Appendix]. However, to within a numerical factor, the damping is described by the last term in Eq. (3.7), where the energy integral C_2 [see Eq. (3.8)] is truncated at distances $x, y \approx \omega_n / 2T \ll 1$. This procedure gives

$$\gamma_k \approx \frac{3b\eta^6}{2^7 \pi^3 S^3} \frac{T^2 \omega_k^2}{\omega_E^3}, \quad (3.11)$$

which is again in agreement with the hydrodynamic theory. However, a comparison of the expression (3.11) with the fluctuation damping (3.2) shows that the hydrodynamic behavior of nuclear spin waves occurs only in a fairly small part of the phase space

$$k \ll \tilde{k} \sim \left(\frac{\omega_n}{\omega_E} \right)^{5/4} \left(\frac{\omega_n}{T} \right)^{1/4} k_{max}. \quad (3.12)$$

The damping of nuclear spin waves with a small dynamic frequency shift can be calculated using the scattering amplitude (2.17). In the case of long-wavelength nuclear spin waves ($k \ll k_0$) the problem simplifies because the damping decrement is dominated by a logarithmically large contribution of the processes of the scattering of these long-wavelength nuclear spin waves by short-wavelength waves ($p, q \gg k_0$), as described in Refs. 9 and 13. The result is

$$\begin{aligned} \gamma_k &= \frac{\eta^6}{3 \cdot 2^5 \pi^3 S} \frac{\epsilon_k T^2}{b \omega_E^5} (\ln A + \text{const}), \\ A &= \min \left\{ \frac{k_0}{k}, \left(\frac{k_{max}}{k_0} \right)^3 \right\}. \end{aligned} \quad (3.13)$$

An increase in the wave vector of a given nuclear spin wave reduces the logarithm in Eq. (3.13). It follows that if $k \gtrsim k_0$, then nuclear spin waves with comparable wave vectors participate in the scattering process. The complexity of the scattering surface makes it impossible to solve the problem analytically. Apart from a numerical factor, the damping result is given by Eq. (3.13) without a logarithmic term.⁹⁾

Comparing the damping of nuclear spin waves due to the processes of four-wave scattering [Eq. (3.13)] with the fluctuation damping, we can demonstrate that the two types of process are of the same order of magnitude if the wave vector of a nuclear magnon is at the limit of the range $k_1^* \leq k \leq k_2^*$ [see Eq. (1.9)]. As pointed out in the Introduction, a spin-wave picture then ceases to be valid. Well inside the spin-wave region when $T \gg \omega_n$, the dominant process is

the fluctuation scattering. The ratio γ_{4n}/γ_{fl} is minimal for $k \approx k_0$, when it is of the order of

$$\frac{\gamma_{4n}}{\gamma_{fl}} \sim \left(\frac{T}{T^*} \right)^2 \sim \left(\frac{r_2^*}{r_0} \right)^3. \quad (3.14)$$

We shall now consider the process of renormalization of the spectrum of nuclear spin waves because of their interaction. At temperatures $T \ll \omega_n$ such renormalization is governed by single-loop diagrams corresponding to the four-wave scattering. If $\omega_0 \ll T$, $\omega_k \ll \omega_n$, the result calculated in the first order in the scattering amplitude (2.18) is of the form

$$\frac{\delta\omega_k}{\omega_k} = -\frac{b^2 \eta^3}{3 \cdot 2^2 S^2} \left(\frac{T}{\omega_E} \right)^2 \left(\frac{\tilde{\omega}_0}{\omega_k} \right)^2 \left(\frac{\omega_E}{\omega_n} \right)^{1/2} \times \left[1 - \frac{3\pi^2}{5} \left(\frac{T\omega_k}{\omega_n \tilde{\omega}_0} \right)^2 \right]. \quad (3.15)$$

Renormalization of the spectrum of short-wavelength nuclear spin waves with energies $\omega_k \approx \omega_n$ is due to their scattering by long-wavelength nuclear spin waves and if $T \gg \omega_0$, we have

$$\frac{\delta\omega_n}{\omega_n} = \frac{\pi^2 b^2 \eta^3}{15 \cdot 2^2 S^2} \left(\frac{T}{\omega_E} \right)^4 \left(\frac{\omega_E}{\omega_n} \right)^{1/2}. \quad (3.16)$$

At temperatures $T \gg \omega_n$, the dominant contribution to the renormalization of the energy of nuclear spin waves is made by the fluctuation process. A calculation of the real part of the diagram in Fig. 6 gives

$$\frac{\delta\omega_k}{\omega_n} = -\frac{\eta^3}{8\pi S} \frac{T\epsilon_0}{\omega_E^2}. \quad (3.17)$$

4. INFLUENCE OF THE DIPOLE-DIPOLE INTERACTION¹⁰⁾ ON THE SPECTRUM AND DAMPING OF NUCLEAR SPIN WAVES

In considering the dynamic properties of the nuclear subsystem of an antiferromagnet we have ignored so far the dipole-dipole interaction. In fact, in antiferromagnets this interaction is exchange-weakened and its contribution to the various quantities is determined by a small parameter $2\pi/\delta$ (Refs. 17 and 18). However, the dipole-dipole interaction gives rise to a qualitatively important effect: it lifts the degeneracy of the spectrum of electron and nuclear spin waves in respect of the wave-vector direction. Consequently, the damping decrement of the excitations becomes anisotropic and a nonzero contribution appears in the damping in the limit $k \rightarrow 0$ and it is due to the fluctuation mechanism. These two circumstances alter considerably the relaxation of nuclear spin waves in the long-wavelength part of the spectrum (see also the Introduction).

In a theoretical analysis we shall allow for the dipole-dipole interaction using perturbation theory and the parameter $2\pi/\delta$ (Ref. 19). The influence of the dipole-dipole interaction on the nuclear subsystem of an antiferromagnet reduces to the renormalization of the Green magnon functions and of the amplitudes of the hyperfine interaction, so that the coefficients V_1 , U_{12} , and W_{1234} of the effective Hamiltonian (2.1) acquire additional—compared with Eqs. (2.2)–(2.4)—factors

$$\left(1 - \frac{(sk_d)^2}{\epsilon_{k_d}^2} \cos^2 \theta_{k_d} \right) \quad (4.1)$$

per each dashed line in Fig. 1. A characteristic wave vector k_d is given by Eq. (1.1) and the angle θ is measured from the z axis which coincides with the spontaneous magnetization direction. Correspondingly the spectrum of nuclear spin waves obtained using (4.1) becomes

$$\omega_k = \omega_n \left[1 - \frac{2bJ_0\omega_n}{\epsilon_0^2} \left(1 - \frac{(sk_d)^2}{\epsilon_0^2} \cos^2 \theta_k \right) \right]^{1/2}, \quad (4.2)$$

whereas the amplitude of the fluctuation scattering is given by the expression

$$\Phi_{fl}(k) = \frac{\omega_k}{4b} \left[1 - \left(\frac{\omega_k}{\omega_n} \right)^2 \right]. \quad (4.3)$$

It should be pointed out that the simplistic allowance for the dipole-dipole interaction by substitution in Eq. (1.5) of the correct electron magnon spectrum ϵ_k obtained including the dipole-dipole interaction gives incorrect results. We find then that the expression for the spectrum of nuclear spin waves given by (4.2) acquires additional “parasitic” terms of the order of $2\pi/\delta$ and these do not disappear for the zero skew angle of the magnetic moments of the electron sublattices.

Using the expressions (4.2) and (4.3), we can calculate the fluctuation damping of nuclear spin waves employing a formula similar to Eq. (3.1). In the range $k \ll k_0$, where the spectrum of nuclear spin waves is typical of an antiferromagnet

$$\begin{aligned} \omega_k &= [\omega_0^2 + v^2(k^2 + k_d^2 \cos^2 \theta_k)]^{1/2}, \\ \omega_0^2 &= \omega_n^2 \frac{H(H+H_D)}{\epsilon_0^2}, \quad v^2 = \frac{2bJ_0\omega_n^3}{\epsilon_0^4}, \end{aligned} \quad (4.4)$$

the fluctuation damping is given by the formula

$$\gamma_{fl}(k) = \frac{\eta^3}{8\pi S} \frac{b'}{b} \frac{\omega_k^3 sk}{\omega_n \omega_E^2} I, \quad (4.5)$$

where for $k > k_d \sin \theta_k$, we have

$$I = \frac{1}{2kk_d} \left[k_d(k^2 - k_d^2 \sin^2 \theta_k)^{1/2} + (k^2 + k_d^2 \cos^2 \theta_k) \arccos \left(\frac{k^2 - k_d^2 \sin^2 \theta_k}{k^2 + k_d^2 \cos^2 \theta_k} \right)^{1/2} \right],$$

whereas for $k < k_d \sin \theta_k$, we obtain

$$I = \frac{\pi}{4kk_d} (k^2 + k_d^2 \cos^2 \theta_k).$$

In the limit $k \rightarrow 0$, Eq. (4.5) reduces to

$$\gamma_{fl}(\theta_k) = \frac{\eta^3}{32S} \frac{b'}{b} \frac{[\omega_0^2 + v^2 k_d^2 \cos^2 \theta_k]^{1/2} sk_d \cos^2 \theta_k}{\omega_E^2 \omega_n}. \quad (4.6)$$

In the range $k \gg k_d$ we shall give a more general result than the expansion of Eq. (4.5) and this result is valid also when $k \gtrsim k_0$:

$$\begin{aligned} \gamma_{fl}(k) &= \frac{\eta^3}{8\pi S} \frac{b'}{b} \frac{\omega_k^3}{\omega_n} \frac{sk}{\omega_E^2} \\ &\times \left\{ 1 + \frac{k_d^2}{2k^2} \left[\cos^2 \theta_k \left(1 + \frac{3k}{\omega_k} \frac{\partial \omega_k}{\partial k} \right) - \frac{1}{3} \right] \right\}. \end{aligned} \quad (4.7)$$

We can show that Eqs. (4.5) and (4.7) match in the range $k_d \ll k \ll k_0$.

5. SMALL PARAMETERS OF THE THEORY AND ESTIMATES OF THE IGNORED CONTRIBUTIONS

In discussing the processes of relaxation of nuclear spin waves at high temperatures we have shown above that the occurrence of large occupation numbers of nuclear spin waves $n(\omega_k/T) \gg 1$ does not by itself result in a divergence of the perturbation theory series in respect of the number of loops in the diagrams. Rigorous calculations including "paramagnetic" diagrams give a factor $(T/T^*)^2$, where T^* is given by Eq. (1.7), which applies to each integration loop. This ensures divergence of the perturbation theory series in the spin-wave range of temperatures $T \lesssim T^*$ (see also the Introduction). The second parameter of the theory is a small quantity $\alpha^2 = \omega_n^2/\epsilon_0\omega_E$ which occurs in Eq. (1.6) for the second moment of the nuclear level. In this connection all the diagrams governing the physical characteristics of the nuclear subsystem can be divided into two classes.

The first class consists of the diagrams containing oriented loops with integration over the momenta not linked by the law of conservation of energy. Examples of such diagrams are given in Fig. 8. All these diagrams make contributions which are small in respect of the parameter α and, therefore, were ignored in the investigated range of temperatures $T \gg \omega_n$ [however, it should be noted that if $T \ll \omega_n$, the graph in Fig. 8a dominates the renormalization of the spectrum of nuclear spin waves given by Eqs. (3.15) and (3.16)]. In particular, the diagrams in Figs. 8a and 8b yield relative corrections to the spectrum of nuclear spin waves of the order of α^2 and $(T^*/T)\alpha^3$, respectively, whereas the diagram in Fig. 8c renormalizes the longitudinal Green function (equal in the first approximation to 2b') by an amount of the order of α^2 and the diagram of Fig. 8d for the correction to the fluctuation vertex has a relative order of smallness $(T/T^*)\alpha$. The diagram in Fig. 8e for the correction to the amplitude of the four-wave scattering includes small contributions of all three types.

All the remaining diagrams belong to the second class. These are, for example, the diagrams shown in Figs. 6 and 7, as well as those given in Fig. 9. The contributions due to these diagrams are governed by the parameter $(T/T^*)^2$. In particular, the diagram in Fig. 9a is a contribution to the damping of nuclear spin waves which is of the same order as the four-wave process [Eq. (3.13) without the logarithmic term]. It is the diagrams of the second class, which become of the same order of magnitude relative to one another, that destroy the spin-wave picture of the nuclear subsystem at $T \sim T^*$. One may expect that well within the paramagnetic

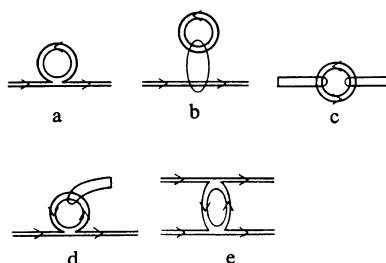


FIG. 8. Diagrams making contributions small in the parameter α .

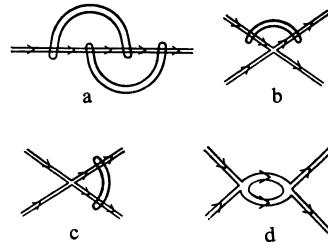


FIG. 9. Diagrams making contributions governed by the parameter $(T/T^*)^2$.

region ($T \gg T^*$) the universal small parameter of the theory is α .

6. CONCLUSIONS

The results obtained in the present study show that the picture of relaxation in the nuclear subsystem of an antiferromagnet changes considerably in the case of a large dynamic frequency shift, compared with the case of a small shift (Refs. 6, 8, 9, and 13).

Firstly, the damping of nuclear spin waves due to the scattering by thermal fluctuations of the longitudinal component of the nuclear spins, which is the main source of relaxation at temperatures $T \gtrsim \omega_n$, acquires an additional frequency dependence [see Eqs. (1.8) and (3.2)] and decreases significantly on increase in the dynamic frequency shift. In the limiting case when the spectrum of nuclear spin waves becomes of the Goldstone type ($\omega_0 = 0$) a hydrodynamic range $k \lesssim \bar{k}$ appears in the long-wavelength part of the phase space of the nuclear subsystems [see Eq. (3.12)] and in this range the damping of nuclear spin waves is governed by the four-wave scattering processes. This behavior of the damping decrements ($\gamma_{lf} \propto k^4$, $\gamma_{4n} \propto k^2$) is due to the interference of the contributions from various terms of the Hamiltonian of the nuclear subsystem (2.1), giving rise to considerable reductions in the scattering amplitudes and to fulfillment of the Adler principle.

Secondly, in the range $T \ll \omega_n$ where thermal fluctuations are weak, the dynamics of the nuclear subsystem is dominated throughout the phase space by the four-wave scattering processes. It is worth pointing out a remarkable feature. If ignoring the interaction with the nuclear subsystem the spectrum of electron magnons in an antiferromagnet becomes of the Goldstone (or almost-Goldstone) type, then when an allowance is made for this interaction in the electron spectrum, a gap appears because of the static action of the nuclei and the Goldstone (or almost-Goldstone) properties are transferred to the nuclear subsystem. All the dependences of the damping decrement of nuclear spin waves on the wave factor and temperature in the "antiferromagnetic" part of the spectrum at $T \ll \omega_n$ repeat, apart from the coefficients, the dependences typical of an easy-plane antiferromagnet or ferromagnet. For example, in the presence of an energy gap in the long-wavelength limit the damping of nuclear spin waves has the form $\gamma \propto T^2 \tilde{\omega}_0^4 \omega_k^{-1}$ [the main contribution is governed by the "gap" term in the amplitude (2.18)]. In the case of "suprathermal" nuclear spin waves ($\omega_k \gg T$), we find that $\gamma_k \propto \omega_k^5 T^4$.

It should also be pointed out that an allowance for the dipole-dipole interaction gives rise to an anisotropy of the damping decrement of nuclear spinwaves in the \mathbf{k} space [see Eqs. (4.5) and (4.7)]. This circumstance is important in discussing the properties of parametrically excited nuclear magnons.

We shall now compare our results with the experimental data. In the case of parametric excitation of nuclear spin waves in various crystals it has been found^{7,20} that the dependence of the damping decrement on the wave vector and temperature is of the type

$$\gamma \propto kT. \quad (6.1)$$

It is natural to attribute such a dependence to the fluctuation mechanism of the scattering of nuclear spin waves. In the experiments carried out at frequencies $\omega_k \rightarrow \omega_n$, the damping decrement is identical with a numerical estimate obtained from Eq. (1.8). The possibility of a strong dependence of this decrement on the frequency of nuclear spin waves is pointed out in Ref. 8. An attempt made in Ref. 20 to detect a strong reduction in the damping decrement of nuclear spin waves in the presence of a large dynamic frequency shift was not successful. Instead of the theoretically predicted dependence $\gamma \propto \omega_k^3$, a characteristic trough-like dependence with a minimum at frequencies 400–500 MHz was observed. In our opinion, this behavior of the damping decrement is due to the imperfections of the investigated samples. An analysis shows²¹ that the process of scattering of nuclear spin waves on fluctuations of the longitudinal component of the spin of a paramagnetic impurity makes a contribution to the damping decrement that decreases on increase in the frequency:

$$\gamma \propto ckT \left(1 - \frac{\omega_k^2}{\omega_n^2}\right)^2 \omega_k^{-1}, \quad (6.2)$$

where c is the paramagnetic impurity density. A similar frequency dependence results from the scattering of nuclear spin waves on crystal structure defects (which may be point or linear).²²

The sum of the expressions (3.2) and (6.2) can account fully for the experimentally observed frequency dependence of the damping decrement of nuclear spin waves. The validity of this assumption confirms also the results of Ref. 23, where a reduction in γ on increase in ω_k is observed in accordance with Eq. (6.2). We can identify the dependence $\gamma \propto \omega_k^3$ in the damping of nuclear magnons by experiments on crystals with controlled amounts of impurities. An extension of the investigated frequency range will also help to give information on the pattern of relaxation of nuclear spin waves characterized by a large dynamic frequency shift.

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APPENDIX

We shall now give the expression for the amplitude of the four-wave scattering of nuclear spin waves in the case of an arbitrary dynamic frequency shift:

$$\Phi_{4n} = -\frac{b\omega_n^2}{(\omega_1\omega_2\omega_3\omega_4)^{1/2}} \left\{ -24bW_{1234} + U_{12} + U_{34} + U_{13} + U_{24} + U_{14} + U_{23} \right\}$$

$$+ \frac{1}{\omega_n^2} [-U_{12}\omega_3\omega_4 - U_{34}\omega_1\omega_2 + U_{13}\omega_2\omega_4 + U_{24}\omega_1\omega_3 + U_{14}\omega_2\omega_3 + U_{23}\omega_1\omega_4] \\ + \frac{1}{4b} [V_1 + V_2 + V_3 + V_4] + \frac{1}{8b^2\omega_n^3} [\omega_n^4 - \omega_1\omega_2\omega_3\omega_4] \right\}.$$

¹When only the magnetoelastic interaction is included, it is found that $\Delta^2 = 2\omega_E\omega_{mes}$, where ω_{mes} is the characteristic energy of the magnetoelastic interaction. A magnetic field H is applied along the direction of easy magnetization in the basal plane.

²The different boson representations of the spin operators, such as the Holstein-Primakoff representation with the $I \rightarrow b$ substitution are incorrect at temperatures $T \gtrsim \omega_n$, when the quantity b differs considerably from I . Although these representations make it possible to obtain the correct spectrum of noninteracting nuclear spin waves of the (1.5) type, the application of these representations to the processes of interaction in the nuclear subsystem requires certain artificial assumptions (see Ref. 8).

³This result does not confirm the *a priori* statements found in the literature that the expansion parameter is in this case the “reciprocal interaction radius” (a/r_0)³.

⁴The inequality $k_d \ll k_0$ is satisfied in real cases.

⁵This description is valid if we ignore the problem of the normal positions of the operators m^α in the Hamiltonian, i.e., if we ignore corrections of the order of $b^{-1}(a/r_0)^3$. These corrections are small both for $T \ll \omega_n$ and $T \gg \omega_n$, in which case their order of smallness is given by $(T/T^*)^2(\omega_n/T)$.

⁶The original variant of Ref. 10 utilizing renormalization of the interaction instead of introduction of Green spin-wave functions is less convenient.

⁷This circumstance is related to the form of the nuclear spin wave spectrum and it slows down considerably the process of relaxation of the nuclear subsystem to a thermodynamic equilibrium state after the excitation of short-wavelength nuclear spin waves.

⁸When “paramagnetic” diagrams are ignored (as is done in the case when the boson representations are used), additional factors of the order of $(k_{max}/k_0)^3$ appear in the expressions for the physical quantities.

⁹Since at high temperatures the damping decrement of short-wavelength ($k \gg k_0$) nuclear spin waves is dominated by the short-wavelength part of the phase space where the dynamic frequency shift is small, this result is valid even if the shift is large in the long-wavelength part.

¹⁰We shall consider only the dipole-dipole interaction between electron spins. The dipole-dipole interactions associated with the magnetic moment of the nuclei can be ignored because of the smallness of the nuclear magneton.

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