The emergence of magnetic order in a nuclear spin system during adiabatic demagnetization

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The transition to the magnetically ordered state during adiabatic demagnetization of a nuclear spin system of a solid dielectric with large initial spin polarization is considered. It is shown that within the framework of the spin-wave description the transition to the state with tilted magnetization can be interpreted as a Bose condensation of magnons. Phase transition points are determined and the types of emergent magnetic structures are identified and investigated. © *1994 American Institute of Physics.*

1. INTRODUCTION

One of the most interesting phenomena taking place in the nuclear spin system of a solid diamagnetic dielectric is the transition to magnetically order states during the cooling of the nuclear subsystem to spin temperatures comparable with the magnitude of the internuclear dipole-dipole interactions.[1] Such cooling takes place in two steps. First, the sample, which is brought to a temperature of ~1 K in a strong external magnetic field, is subjected to dynamic polarization of its nuclei,[1,2] during which polarization is transferred from the electronic spins to the nuclear spins. By this process a nuclear spin polarization close to 1 is attained. Then a weak rf field is turned one, whose frequency is varied smoothly from a grossly nonresonant value to the Larmor precession frequency of the nuclear spins. As a result, the magnitude of the effective field in the rotating frame slowly decreases in time. When the effective field becomes comparable with the internal local dipole fields, a transition takes place from ordering in the external field to ordering in the local fields, i.e., to the emergence of magnetically ordered structures. A description of such a process, as well as the types of emergent magnetic structures, is the goal of the present paper.

Despite the smallness of the nuclear magnetic moments and the associated experimental difficulties in obtaining large nuclear polarizations, besides practical applications (see Ref. 2, polarized targets) there are other reasons making nuclear magnetism an extraordinarily interesting object of study. These consist in the substantial differences between nuclear and magnetic magnets. For light nuclei (^{1}H, ^{10}F), in a diamagnetic dielectric practically the only spin interaction is the dipole-dipole, for which the Hamiltonian is known exactly and does not contain any empirical parameters. At low temperatures the spin degrees of freedom are maximally isolated from the other degrees of freedom (those of the lattice). For example, for the materials of polarized targets the typical relaxation rate of nuclear demagnetization is of the order of 10^{-3} s^{-1}. The system can easily be cooled to low negative as well as low positive spin temperatures. In addition, the dipole-dipole interaction is long-range, in contrast to the primary interaction in electronic magnets—the exchange interaction, which leads to important features in the spectrum of one-particle excitations.

In contrast to electronic antiferromagnetics[3] or an exciton gas,[4] when studying the quantum peculiarities in a nuclear magnet associated with the formation of a bose condensate it is impossible to improve the agreement between experiment and theory by fitting the form and constants of the interaction, and the indicated peculiarities of the dipole-dipole interactions to new effects[1] which are absent from electronic spin systems.

2. THE SPIN-WAVE REPRESENTATION OF THE HAMILTONIAN

We will consider a system of N nuclear spins 1/2, located at the nodes of a simple cubic lattice. The Hamiltonian in a strong external field H_0 has the form

$$\mathscr{H}=-\omega_0 S_z+\mathscr{H}_{dz}, \tag{1}$$

where $\omega_0=\gamma H_0$, where γ is the gyromagnetic ratio and $\mathscr{H}_{dz}$is the secular (relative to the z axis) part of the dipole-dipole interaction:

$$\mathscr{H}_{dz}=\frac{1}{2}\sum_{l,l'}^{N} b_{l,l'}(3S_l^zS_{l'}^z-\mathbf{S}_l\mathbf{S}_{l'}),$$

$$b_{l,l'}=\gamma^2\hbar^2\frac{1-3\cos^2\theta_{l,l'}}{2|\mathbf{l}'-\mathbf{l}|^3}, \tag{2}$$

where $\theta_{l,l'}$ is the angle between the vector $\delta=\mathbf{l}'-\mathbf{l}$ and the z axis.

At large values of the nuclear polarization $p(1-|p|\ll 1)$ we can replace (2) by the Hamiltonian of the rarefied magnon gas.[5,6] This is achieved with the help of the Dyson–Maleev transformation:[7]

$$S_l^+=a_l,\quad S_l^-=a_l^+-a_l^+a_l^+a_l,\quad S_l^z=\tfrac{1}{2}-a_l^+a_l \tag{3}$$

followed by the Fourier transform

$$a_k^+=\frac{1}{\sqrt{N}}\sum_l a_l^+\exp(-i\mathbf{kl}),\quad b_k=\sum_\sigma b_\sigma\exp(-i\mathbf{k}\delta). \tag{4}$$

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As a result, accurate to within a small constant term, we obtain the following representation for $\mathscr{H}$:

$$\mathscr{H}=\sum_k \varepsilon_k a_k^+ a_k+\frac{1}{4N}\sum_{k,p,q} I_{k,p}^q a_{k-q}^+ a_{p+q}^+ a_k a_p, \qquad (5)$$

where the one-magnon excitation spectrum has the form

$$\varepsilon_k=\omega_0-b_0-\tfrac{1}{2}b_k=\omega_0-b_0+\tilde{\varepsilon}_k, \qquad (6)$$

and the amplitude of the magnon-magnon interaction is equal to

$$I_{k,p}^q=b_{p+q}+b_{q-k}+2b_q+2b_{p+q-k}. \qquad (7)$$

The magnon creation and annihilation operators a_k^+ and a_k obey the commutation rules for bosons. In the spectrum (6) b_0 is the frequency shift due to the demagnetizing field. In what follows we will assume that the sample has a spherical shape, so that for a cubic lattice $b_0=0$.

3. THE MAGNON SPECTRUM

The spectrum of one-magnon excitations (6) $\tilde{\varepsilon}_k=b_k/2$ is highly anisotropic and depends on the orientation of the crystallographic axes with respect to the external field. It contains two qualitatively different contributions: one from the immediate environment, and the other from large distances, associated with the long-range nature of the dipole-dipole interaction. For example, in the orientation $\mathbf{H}_0\|[001]$, the contribution from nearest neighbors is

$$\tilde{\varepsilon}_k^{(1)}=\frac{\gamma^2\hbar}{2a^3}(2\cos k_z a-\cos k_x a-\cos k_y a), \qquad (8)$$

where a is the lattice constant. Here there is a saddle point at $k=0$, and the maximum and minimum of $\tilde{\varepsilon}_k^{(1)}$ are realized on the boundary of the Brillouin zone. The long-range contribution is obtained by replacing the sum in the expression for b_k (4) by an integral and has the form[1,8]

$$\tilde{\varepsilon}_k^{(2)}=-\left(\frac{\gamma^2}{4a^3}\right)\frac{4\pi}{3}(3\cos^2\varphi-1), \quad \mathbf{k}\to 0, \qquad (9)$$

where φ is the angle between $\mathbf{k}$ and the external field. This contribution, naturally, does not depend on the orientation of the crystal and has a singular form: the value of $\tilde{\varepsilon}_k^{(2)}$ as $\mathbf{k}\to 0$ depends on the direction by which $\mathbf{k}$ tends to 0.

Thus, qualitatively the spectrum $\tilde{\varepsilon}_k$ can be considered the sum of the two terms (8) and (9). The spectrum $\tilde{\varepsilon}_k$ can be obtained more accurately by numerical calculation.[8] We present here only the positions of the maxima and minima of $\tilde{\varepsilon}_k=\varepsilon_k-\omega_0$ for three orientations of the crystal (in units of $\gamma^2\hbar/4a^3$, $\mathbf{k}^*$ is the value of the wave vector for which a maximum or minimum value of $\tilde{\varepsilon}_k$ is realized):

1) $\mathbf{H}_0\|[001]$

$$\tilde{\varepsilon}_k^{\max}=5.352, \quad \mathbf{k}^*=\left(\frac{\pi}{a},\frac{\pi}{a},0\right),$$

$$\tilde{\varepsilon}_k^{\min}=-9.687, \quad \mathbf{k}^*=\left(0,0,\frac{\pi}{a}\right).$$

2) $\mathbf{H}_0\|[110]$

$$\tilde{\varepsilon}_k^{\max}=4.843, \quad \mathbf{k}^*=\left(0,0,\frac{\pi}{a}\right),$$

$$\tilde{\varepsilon}_k^{\min}=-8\pi/3, \quad \mathbf{k}^*\to 0, \quad \mathbf{k}^*\|\mathbf{H}_0.$$

3) $\mathbf{H}_0\|[111]$

$$\tilde{\varepsilon}_k^{\max}=4\pi/3, \quad \mathbf{k}^*\to 0, \quad \mathbf{k}^*\perp\mathbf{H}_0,$$

$$\tilde{\varepsilon}_k^{\min}=-8\pi/3, \quad \mathbf{k}^*\to 0, \quad \mathbf{k}^*\|\mathbf{H}_0.$$

We see that in the first case the contribution of the immediate environment is large, and it is this term that determines the regular maximum and minimum on the boundary of the Brillouin zone. In the third case the contribution from the nearest neighbors vanishes, and the extrema are determined by the long-range singular contribution. The second orientation is intermediate in this sense.

4. ADIABATIC DEMAGNETIZATION

In the absence of an rf field the equilibrium magnon distribution is established in the system:

$$n_k=\langle a_k^+ a_k\rangle=\frac{1}{e^{\beta(\varepsilon_k-\mu)}-1}. \qquad (10)$$

The possibility of the chemical potential μ taking values different from zero is due to the presence of two independent additive integers of the motion: the number of particles (the z-components of the total magnetization) and the energy. The equilibrium density matrix thus depends on two independent thermodynamic parameters. In the description in terms of particles, it is convenient to choose as such parameters the inverse temperature β and the chemical potential μ.

The polarization along the z axis is determined by the total number of magnons n

$$p=1-2n/N=1-\frac{2}{N}\sum_k n_k, \qquad (11)$$

and the main contribution to the energy for $1-|p|\ll 1$ ($n\ll N$) comes from the sum of the single-particle energies

$$E\approx\sum_k \varepsilon_k n_k. \qquad (12)$$

In the absence of an rf field the Hamiltonian of the system and consequently the equilibrium density matrix are invariant to rotations about the z axis. This means that the equilibrium magnon numbers (10) do not vary when we consider the problem from a rotating coordinate system rotating with frequency ω, to which we can transform via the unitary transformation

$$U(t)=\exp\{-i\omega t S_z\}=\exp\left\{-i\omega t\left(\frac{N}{2}-\sum_k a_k^+ a_k\right)\right\}. \qquad (13)$$

The magnon energy in the rotating frame ε_k' is shifted relative to its value in the laboratory frame by the amount ω:

$$\varepsilon_k'=\varepsilon_k-\omega. \qquad (14)$$

From conservation of magnon number n_k we obtain

$$\varepsilon_k-\mu=\varepsilon_k'-\mu'=\varepsilon_k-\omega-\mu', \tag{15}$$

where μ' is the chemical potential in the rotating frame. Turning on a weak rf field leads to the result that β and μ begin to vary slowly in time and via a succession of quasi-equilibrium states the system tends to arrive at a new equilibrium state. Total equilibrium corresponds to vanishing of the chemical potential in the rotating frame, where the Hamiltonian does not depend on time. Thus, for example, if the initial state corresponded to total equilibrium in the laboratory frame $\mu=0$, $\mu'=-\omega$, then with saturation of the line the system should arrive at the state with $\mu'=0$.

In can be shown that such a state cannot be reached by saturation at a frequency within the spectrum ε_k since, according to Eq. (10), negative values should obtain for some n_k (for $\varepsilon_k-\omega\langle 0,\beta\rangle 0$). This apparent contradiction is eliminated if we take into account that growth of the number of magnons (i.e., of the chemical potential) is also accompanied by a shift of the boundary of the spectrum due to increase of the interaction between the magnons.[5,6] In the case of low temperatures ($\beta\omega_{\rm loc}\gg 1$, $\omega_{\rm loc}\approx\varepsilon_k^{\max}-\varepsilon_k^{\min}$) this shift can be represented as:[5,6]

$$\Delta\varepsilon_k^{\min}=2(1-p)(\omega_0-\varepsilon_k^{\min})=-2(1-p)\tilde{\varepsilon}_k^{\min}. \tag{16}$$

Thus, saturation is complete when the lower boundary of the spectrum rises (or the upper boundary drops for the case of negative energies) so much that the energy of the rf field coincides with the boundary of the spectrum. In this case the final polarization is determined by Eq. (16), and the energy, by the law of conservation of energy in the rotating frame.

Absorption of a nonresonant quantum of the rf field is a complicated process which is realized by interaction between the magnons (in the absence of interaction, e.g., in the limit $n\to 0$ $(p\to 1)$, the shape of the absorption lines is a δ-function at the Larmor precession frequency. However, in the case of a weak rf field, when thermodynamic equilibrium is possible, the description turns out to be quite simple: absorption of a quantum of the rf field is equivalent to adding one magnon with energy equal to the frequency of the field. Here we assume that the deviation from the equilibrium distribution is always small.

Let us now consider the most effective means of cooling a spin system—adiabatic demagnetization in the rotating frame. For definiteness, we will consider the case of positive temperatures, when the process begins with $\omega<\omega_k^{\min}$, and then the frequency of the rf field ω is slowly increased. This process is most simply described in a coordinate system corotating at frequency ω. If the energy is reckoned from the bottom of the magnon energy band, taking account of the shift (16), then this process looks like the addition of magnons with zero energy, i.e., the energy in the rotating frame is conserved. Increase in the magnon number obtained from the condition that the shift (16) is tuned to the frequency of the rf field $\varepsilon_k^{\min}+\Delta\varepsilon_k^{\min}=\omega$.

Let us assume that we have stopped the process of adiabatic demagnetization at some frequency ω^* such that the polarization is still large ($\omega^*-\varepsilon_k^{\min}\ll\omega_0-\varepsilon_k^{\min}=-\tilde{\varepsilon}_k^{\min}$). If now we measure the NMR absorption signal with the help of a probe field (that is, we measure the imaginary part of the susceptibility), then since the equilibrium state thus obtained will correspond to $\mu=0$ with frequency ω^*, for $\omega<\omega^*$ in the frame rotating at frequency ω, the chemical potential $\mu>0$ and the magnon number decays, but for $\omega>\omega^*$, the chemical potential $\mu<0$ and the magnon number grows. That is, as in the high-temperature case,[9] in that part of the line that has already been passed through, absorption is replaced by emission, and the line intensity vanishes at $\omega=\omega^*$.

5. BOSE CONDENSATION OF MAGNONS AND FORMATION OF ORDERED STRUCTURES

As was stated in the previous section, adiabatic demagnetization in the corotating frame is described as a process in which the magnon number increases while the energy stays fixed. At some critical magnon density n_c/N Bose condensation takes place in the system. The possibility of Bose condensation in a magnon gas for various spin systems was previously considered in Refs. 3, 6, and 10–12. The transition point is easiest to find if we substitute the equilibrium distribution (10) with $\mu=0$ (the energy is reckoned from the bottom of the band taking account of the shift (16) in the expressions for the particle number (11) and the energy (12). If we eliminate β from these equations, we obtain a relation for $n_c(E)$. For $n>n_c$, n_0 of the "excess" magnons show up in the condensate: $n=n_c+n_0$. Thus, during adiabatic demagnetization, growth of the magnon number is at first accompanied by a lowering of the temperature, but after the transition point the temperature ceases to vary and growth of the magnon number is due to an increase of their number in the condensate. Since energy is conserved in the tracking rotating frame, in the capacity of the energy in $n_c(E)$ we should substitute the initial energy E_0. Taking into account the fact that the initial state can be taken to be a high-temperature state (for it $n_k=n/N$ and $\Delta\varepsilon_k^{\min}=0$, Ref. 5), we obtain from the condition $\Sigma_k\tilde{\varepsilon}_k=0$

$$E_0=-n_i\tilde{\varepsilon}_k^{\min}, \tag{17}$$

in the rotating frame, where n_i is the initial magnon number, determined by the initial polarization p_i: $p_i=1-2n_i/N$. The transition point, or equivalently the relation between n_c and n_i, depends on the form of the spectrum near the minimum. For example, for the orientation $\mathbf{H}_0\|[001]$, when there is a regular minimum on the boundary of the Brillouin zone $\mathbf{k}=(0,0,\pi/a)$, the spectrum near the minimum has the form

$$\tilde{\varepsilon}_k\approx-2c_1+c_1A_1(\delta\mathbf{k})^2a^2, \tag{18}$$

where $\delta\mathbf{k}=\mathbf{k}-\mathbf{k}^*$, $A_1=0.31$, $c_1=1.211\gamma^2\hbar/a^3$. In this case

$$\frac{n_c}{N}=\frac{\xi(3/2)}{[3\pi A_1\xi(5/2)]^{3/5}}\left(\frac{n_i}{N}\right)^{3/5}, \tag{19}$$

where $\xi(x)=\Sigma_{m=1}^{\infty}m^{-x}$. This relation follows from the condition that $\beta\omega_{\rm loc}\gg 1$ at the transition point, i.e., the magnons are located near the bottom of the band and it is possible to use expansion (18). As calculation shows, this condition is fulfilled if the polarization p_c at the transition point is large: $1-p_c\ll 1$. Below, within the framework of the mean field approximation, we will see that this same transition can be considered a phase transition ordering the transverse spin

components in the local fields created by them. Since the transverse components are small for $1-p \ll 1$, the transverse fields which they create are much smaller than ω_{loc}. Hence, $\beta\omega_{\text{loc}} \gg 1$ at the transition point. For example, for the experimentally attainable initial polarization $p_i=0.98$, with the help of relation (19) we obtain $p_c=0.85$, i.e., the polarization at the transition point is still very high.

Condensation of magnons to the state with $\mathbf{k}^*$ lying on the boundary of the Brillouin zone means the appearance of antiferromagnetic ordering of the transverse components of the nuclear spins. Condensation to the state with $\mathbf{k}^* \to 0$ should be accompanied by the onset of ferromagnetic ordering. Indeed, in the $\mathbf{H}_0 \| [111]$ orientation at positive temperatures the authors of Ref. 13 experimentally detected the existence of nondecaying transverse magnetization in the rotating frame. It is interesting that the magnetically ordered state is realized in the rotating frame while in the laboratory frame magnetization precesses with the Larmor frequency. In order to find the transition point in this case, we expand the spectrum near the singular minimum $\mathbf{k}^* \to 0$, $\mathbf{k}^* \| \mathbf{H}_0$:

$$\tilde{\varepsilon}_k \approx -2c_2 + c_2(3\varphi^2 + A_2 k^2 a^2), \tag{20}$$

where φ is the angle between $\mathbf{k}$ and $\mathbf{H}_0$, $c_2 = \pi\gamma^2\hbar/3a^3$, $A_2 \approx 1\pi$ (Ref. 8). The dependence of the magnon number at the transition point n_c on the magnon number in the initial state n_i has the form

$$\frac{n_c}{N} = \frac{\xi(5/2)}{96(\pi A_2)^{3/7}} \left[\frac{384}{5\xi(7/2)} \frac{n_i}{N} \right]^{5/7}. \tag{21}$$

Note that for the given orientation ($\mathbf{H}_0 \| [111]$), the possibility of using expansion (20) at the minimum instead of the total spectrum imposes a rather stringent constraint on the magnitude of the initial polarization. This is because the singular minimum is "narrow" [with decreasing temperature, the wave vectors of the occupied states group themselves about the direction $\mathbf{H}_0$; see Eq. (20)] and holds only a small number of magnons (for given temperature and chemical potential), while a little higher in energy there exists a wide regular local minimum on the boundary of the Brillouin zone. A fairly low temperature is needed to get most of the magnons to cluster near the minimum (20). Calculation shows that the analytic expression (21) for the transition point is valid only for initial polarization of at least $p_i \approx 0.99$.

6. THE MEAN FIELD APPROXIMATION

In order to form a qualitative picture of the transitions to the magnetically ordered states, it is useful to consider the case $p_i \to 1$ within the framework of the classical mean field approximation. Let $T = +0$. Then the polarization of each spin is maximal and aligned with the corresponding local field. Let us find the energy E_{xy} of the state with tilted magnetization in the external field Δ. Toward this end, we replace the average values of the spin components by the corresponding values of the longitudinal $P_\parallel$ and transverse $P_\perp$ polarizations:

$$\langle S_l^z \rangle = \tfrac{1}{2} P_\parallel, \quad \langle S_l^x \rangle = \tfrac{1}{2} P_\perp \cos(\mathbf{kl}), \quad \langle S_l^y \rangle = \tfrac{1}{2} P_\perp \sin(\mathbf{kl}). \tag{22}$$

Noting that $b_0 = 0$, we represent the energy in the form

$$E_{xy} = \frac{1}{2} \sum_l (\omega_l^x \langle S_l^x \rangle + \omega_l^y \langle S_l^y \rangle) - N\Delta \frac{1}{2} p_\parallel, \tag{23}$$

where

$$\omega_l^x = -\sum_{l' \neq l} b_{ll'} \langle S_{l'}^x \rangle, \quad \omega_l^y = -\sum_{l' \neq l} b_{ll'} \langle S_{l'}^y \rangle. \tag{24}$$

Substituting expressions (24) into (22) in Eq. (23), we obtain

$$E_{xy} = -\frac{N}{8} b_k P_\perp^2 - \frac{N}{2} \Delta p_\parallel \tag{25}$$

and, thus, the minimum energy per spin for the states with tilted magnetization is given by

$$E_{xy}^{\min} = -\tfrac{1}{8} b_k^{\max} p_\perp^2 - \tfrac{1}{2} \Delta p_\parallel. \tag{26}$$

It can easily be seen that expression (26) is exactly equal to the energy of a boson gas (per anode) at zero temperature in the limit $n \to 0$. In an analogous way we can find the energy of a longitudinal magnet E_z, for which $\langle S_l^x \rangle = \langle S_l^y \rangle \equiv 0$:

$$E_z^{\min} = \tfrac{1}{4} b_k^{\min}. \tag{27}$$

The Zeeman energy here is equal to zero since $\mathbf{k}^* \neq 0$. Comparing energies (26) and (27), we find that the following structures correspond to a minimum in the energy:

1) $\Delta > \Delta_{\text{cr}}^{(1)} = \tfrac{1}{2} b_k^{\max}$, $p_\parallel = 1$;

2) $\Delta_{\text{cr}}^{(1)} > \Delta > \Delta_{\text{cr}}^{(2)}$,

$\cos\theta = \Delta/\Delta_{\text{cr}}^{(1)}$, $(p_\parallel = p\cos\theta,\ p_\perp = p\sin\theta)$:

3) $\Delta < \Delta_{\text{cr}}^{(2)} = \sqrt{\tfrac{1}{2} b_k^{\max}(-b_k^{\min} - \tfrac{1}{2} b_k^{\max})}$, $E_z^{\min} < E_{xy}^{\min}$.

Hence it is clear that in a strong external field all the spins are aligned with it, and for $\Delta = \Delta_{\text{cr}}^{(1)}$ the transition to the state with tilted magnetization begins and the spin tilt angle θ (with respect to the external field) grows from zero at $\Delta = \Delta_{\text{cr}}^{(1)}$ to θ_{cr} at $\Delta = \Delta_{\text{cr}}^{(2)}$, and with further decrease of Δ there takes place a discontinuous transition to the longitudinal structure. For example, for the orientation $\mathbf{H}_0 \| [001]$ $\Delta_{\text{cr}}^{(1)} = 4.844(\gamma^2\hbar/2a^3)$, $\Delta_{\text{cr}}^{(2)} = 1.568(\gamma^2\hbar/2a^3)$, $\cos\theta_{\text{cr}} = 0.324$. For $\Delta < \Delta_{\text{cr}}^{(1)}$ there takes place a transition to the state with tilted magnetization and antiferromagnetic ordering of the transverse components with wave vector $\mathbf{k}^* = (0, 0, \pi/a)$, for $\Delta < \Delta_{\text{cr}}^{(2)}$ the system becomes a longitudinal antiferromagnet with wave vector $\mathbf{k}^* = (\pi/a, \pi/a, 0)$ since it is precisely for this $\mathbf{k}^*$ in the orientation $\mathbf{H}_0 \| [001]$ that the energy (27) is minimum.

Naturally, the tilted state exists only for $\Delta_{\text{cr}}^{(1)} > \Delta_{\text{cr}}^{(2)}$. This condition is equivalent to $b_k^{\max} > |b_k^{\min}|$ or, what is the same, $|\tilde{\varepsilon}_k^{\max}| > \tilde{\varepsilon}_k^{\max}$. For positive temperatures it is fulfilled for all orientations, and, correspondingly, it is violated for negative temperatures, for which the roles of the energy maxima and minima change places.

Thus, for negative temperatures the above approach (Bose condensation of magnons) does not take place since a change in the ground state takes place earlier (at larger detunings).

It is interesting to note that in the limit $p_1 \to 1$ the classical mean field theory gives the correct value of the transition point $\Delta = \Delta_{cr}^{(1)}$, or equivalently $\omega = \varepsilon_k^{min}$, i.e., not only thermal (which is to be expected at $T=0$), but also quantum fluctuations turn out to be negligible. At the same time, for $p<1$ the description of the given transition as a Bose condensation of a weakly nonideal magnon gas when only the diagonal part of the interactions, which causes the level shifts, is taken into account is more accurate then the mean field approximation for spins.

The question may arise whether a more accurate account of the interaction between the magnons is needed since it alters the form of the spectrum near the minimum (from quadratic to linear) and can change the transition point and the behavior in its vicinity. We make a simple estimate of the number of magnons n_1 for which the energy of interaction with other magnons $\Delta\varepsilon$ is comparable with the eigenenergy. We reckon the momentum $\mathbf{k}$ from $\mathbf{k}^*$, and the energy, from $\varepsilon_k^{min} + \Delta\varepsilon_k^{min}$. We set $\varepsilon_k \approx \omega_{loc}(ka)^2$. Then from the condition $\Delta\varepsilon \approx \omega_{loc} n/N$, $\omega_{loc}(k_1 a)^2 \approx \Delta\varepsilon$, we obtain $(k_1 a)^2 = n/N$, and, consequently, $n_1/N \approx (k_1 a)^2 = (n/N)^{3/2}$. Thus, for $n/N \ll 1$, $n_1 \ll n$, i.e., the fraction of magnons in the region with altered spectrum is small, and their contribution to the thermodynamic functions is also small.

To conclude this section, let us briefly discuss the question of observables. It is easy to see that in the state with the Bose condensate the transverse magnetization (or its Fourier component for $\mathbf{k}^* \neq 0$) is equal to zero, which contradicts the classical interpretation of such a state as a state with tilted magnetization. The reason for this lies in the uncertainty principle: for an exactly fixed longitudinal magnetization (prescribed magnon number) the transversed component is completely indeterminate. This question here acquires significance since in one state (the condensate) there turns out to be a macroscopically larger number of magnons. However, an arbitrarily weak (macroscopically) perturbation leads to the result that the magnon number ceases to be a good quantum number, and the transverse magnetization can take on a value different from zero. To calculate mean values, we can, as is customary in such cases,[14,15] replace the operators a_k^* and a_k^{+*} by the number $n_0^{1/2}$ (see also the discussion of this question in Ref. 3).

7. CONCLUSION

Although the behavior of spin systems at low temperatures is substantially complicated, restricting our consideration to the case of only large polarizations allows us to make a significant advance in the description of the dynamics and thermodynamics of the system. A simplification arises due to the smallness of the transverse spin tilts, when the restrictions on the length of the spin vector become unimportant. This situation is reminiscent of the spherical model of a ferromagnet,[16] the phase transition to which is completely equivalent to a Bose–Einstein condensation.[17] In our approach the simplification is connected with the partial diagonalization of the Hamiltonian, when most of the dipole-dipole interactions is included in the single-particle excitation spectrum.

The representation of a nuclear spin system with the help of a weakly nonideal magnon gas has enabled us to describe the thermodynamics of spin cooling during adiabatic demagnetization, to find the transition points to the magnetically ordered states with tilted magnetization and the types of emergent structures. Along with this, a number of features have been identified in the behavior of nuclear spin systems, for which at present even a qualitative explanation is lacking. This includes, for example, the observation of nonequilibrium states in CaF_2 in the orientation $\mathbf{H}_0 \| [111]$ at positive temperatures,[13] the difference in the disappearance time of structures in LiH when measuring using neutron scattering and NMR,[18] the anomalous behavior of spin-relaxation relaxation.[19] We hope that subsequent application of the spin-wave approach will answer some of these questions.

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