positive $C_+(-\infty, +\infty)$ or negative $C_-(+\infty, -\infty)$ part of the integration contour C. Since all the internal vertices of the diagram are taken both on C_+ and on C_-, and the vertex lying on C_- is assigned the factor -1, the contribution of the diagram can be different from zero only if the extreme right vertex on the time axis is an external vertex (over the index i of which no summation is carried out). If we denote by $P_{ii_1\ldots i_n}(x, x_1, \ldots, x_n)$ the contribution of the diagram with external vertices x, $x_1, \ldots, x_n$, then at $t > \max\{t_1, \ldots, t_n\}$ we have

$$P_{1i_1\ldots i_n}(x, x_1, \ldots, x_n) = -P_{2i_1\ldots i_n}(x, x_1, \ldots, x_n).$$

It follows from this, in particular, that expressions of the type (2.8) and (2.12) are independent of the index i.

Next,

$$\sum_{i_1\ldots i_n=1,2} P_{ii_1\ldots i_n}(x, x_1, \ldots, x_n) = 0,$$

if at least one of the time-dependent arguments $t_1, \ldots, t_n$ exceeds t. From this follows the retarded character of the contribution of any diagram to the kinetic coefficients defined by relations of the type (2.8) and (2.12).

It follows also from the foregoing that the so-called vacuum loops are absent in the Keldysh technique.[6] In fact, the vacuum loop is not connected with any of the external vertices. Consequently, it corresponds to a zero factor. In the same manner it is easy to verify that only connected diagrams contribute to the kinetic coefficients (2.8) and (2.12).

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Establishment of equilibrium between the nuclear and electron subsystems on dynamic cooling of nuclei

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The dynamics of nuclear spins in conditions of nuclear dynamic cooling is investigated. The process of establishment of the stationary polarization is analyzed for various intensities of the alternating magnetic field. An equation describing the nuclear polarization process in the case of a strong saturating field at low temperatures is obtained.

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In recent years the introduction of the concept of a spin–spin interaction reservoir has turned out to be extremely fruitful in the development of magnetic resonance in solids.[1–3] According to this concept the spin–spin interaction energy (more precisely, its secular part) is regarded as a separate energy reservoir, isolated, generally speaking, from the Zeeman energy of the spins in the external magnetic field $\mathbf{H}_0$ and characterized by its own temperature, which, under certain conditions, can differ greatly from the Zeeman temperature.

In Refs. 2 and 3, the existence of thermal contact between the dipole reservoir of paramagnetic impurities and the Zeeman system of the nuclei[4] was predicted theoretically, and was later confirmed in numerous experiments. In the presence of near-resonance saturation of the EPR the temperature of the dipole reservoir of the electron spins is lowered. The presence of the effective coupling with the Zeeman system of the nuclei leads to lowering of the nuclear Zeeman temperature too, and this increases the nuclear polarization. This method of polarization has been named the "method of dynamics cooling of nuclei." A number of theoretical and experimental papers[5] are devoted to the study of this method. The method of dynamic cooling of nuclei

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has been investigated in Refs. 6–8 for the case of an inhomogeneously broadened EPR line at low temperatures, which is the case of greatest interest for the preparation of nuclei of high polarization. The dependence of the stationary nuclear polarization on the lattice temperature and on the alternating magnetic field, obtained in Refs. 6 and 7, is in good agreement with the experiments.[8] In addition, the time for establishment of the stationary polarization of the nuclei, which until now has not been studied theoretically in detail, has been measured in the experiments of Refs. 9 and 10.

In the present work we investigate the dynamics of the nuclear spins in conditions of dynamic nuclear cooling.

The Hamiltonian of a system of electron and nuclear spins placed in a constant magnetic field $\mathbf{H}_0$ and an alternating field $\mathbf{H}_1(t)$ whose frequency Ω is close to the Larmor frequency of the magnetic ions has, in this case, the form

$$\mathcal{H}=\sum_n(\omega_S+\omega_n)S_n^z-\omega_I\sum_i I_i^z+\mathcal{H}_d^{sec}+\mathcal{H}_{IS}+{}^1/_2\omega_1(S^+e^{-i\Omega t}+S^-e^{i\Omega t}), \quad (1)$$

where

$$\mathcal{H}_d^{sec}=\sum_{nm}({}^1/_2A_{nm}S_n^zS_m^z+B_{nm}S_n^+S_m^-),$$

$$\mathcal{H}_{IS}=\sum_{i,n}V_{in}^{zz}S_n^zI_i^z+\frac{1}{2}\sum_{i,n}(V_{in}^{+z}I_i^-+V_{in}^{-z}I_i^+)S_n^z;$$

$$\hbar=1, \quad \omega_1=\gamma_SH_1, \quad S^\pm=\sum_n S_n^\pm.$$

In (1) the first two terms are the Zeeman energies of the electron and nuclear spins, respectively, ω_n is the spread in the Larmor frequency of the inhomogeneously broadened EPR line, $\mathcal{H}_d^{sec}$ is the secular part of the dipole–dipole interaction of the paramagnetic impurities, $\mathcal{H}_{IS}$ is the hyperfine interaction of the nuclear spins with the magnetic ions, and the last term describes the interaction of the alternating field with the electron spins.

In the rotating coordinate frame, in which explicit time dependence is absent and a thermodynamic description of the spin system is therefore possible, the Hamiltonian has the form

$$\mathcal{H}'=\sum_n(\omega_S+\omega_n-\Omega)S_n^z-\omega_I\sum_i I_i^z+\mathcal{H}_d^{sec}+\mathcal{H}_{IS}+{}^1/_2\omega_1(S^++S^-). \quad (2)$$

If the alternating magnetic field is weak ($\omega_1 \ll \omega_d$ where $\omega_d^2=\mathrm{Tr}(\mathcal{H}_d^{sec})^2/\mathrm{Tr}(S^z)^2$) and the spectral diffusion of the inhomogeneously broadened EPR line is fast, then, after a certain short time interval has elapsed, quasi-thermodynamic equilibrium, describable by three parameters—the temperatures of the Zeeman subsystem of the electrons (β_S^{-1}), of the dipole reservoir of the paramagnetic impurities (β_d^{-1}), and of the nuclear Zeeman subsystem (β_I^{-1})—is established in the spin system.

As was done in Ref. 11, it is not difficult to obtain equations in the high-temperature approximation that describe the subsequent evolution of the spin system[2)] that arises from the interaction with the alternating field and from the hyperfine interaction of the electron and nuclear spins:

$$\frac{d\beta_S}{dt}=-W(\omega_S-\Omega)(\beta_S-\beta_d),$$

$$\frac{d\beta_d}{dt}=-\frac{(\omega_S-\Omega)^2}{\omega_d^2}W(\omega_S-\Omega)(\beta_S-\beta_d)-\frac{\beta_d-\beta_I}{T_{dI}},$$

$$\frac{d\beta_I}{dt}=-\frac{\beta_I-\beta_d}{T_{Id}}.$$

Here $W(\omega)=\pi\omega_1^2g(\omega)$ is the probability of spin transitions induced by the alternating field, $g(\omega)$ is the EPR lineshape, and

$$\frac{1}{T_{dI}}=-[\mathrm{Sp}(\mathcal{H}_d^{sec})^2]^{-1}\int_{-\infty}^{0}e^{\varepsilon t}\,\mathrm{Sp}\{[\mathcal{H}_d^{sec},\mathcal{H}_{IS}][\mathcal{H}_I,\mathcal{H}_{IS}(t)]\}\,dt,$$

$$\frac{1}{T_{Id}}=\frac{c_d}{c_I}\frac{1}{T_{dI}}, \quad \mathcal{H}_I=-\omega_I\sum_i I_i^z,$$

where c_I and c_d are the specific heats of the nuclear spins and dipole reservoir, respectively; T_{dI}^{-1} is the rate of relaxation of the dipole reservoir of electrons to the nuclear spin system; T_{Id}^{-1} is the rate of relaxation of the nuclear subsystem to the dipole reservoir of the magnetic ions.

If the saturating field is very weak, i.e., $W<T_{dI}^{-1}$, then, in the first stage, a single temperature for the dipole reservoir of magnetic ions and nuclear spins is established in a time $(1/T_{Id}+1/T_{dI})^{-1}$. Next the spin system tends to a stationary state with rate W. Therefore, the rate of establishment of the stationary nuclear polarization is proportional to the intensity of the alternating field.

In the opposite case, when $W>T_{dI}^{-1}$, a single temperature is first established in the electron system and then the nuclear system comes to equilibrium with the electrons in the time T_{Id}, so that, in this case, the rate of establishment of the stationary nuclear polarization does not depend on the intensity of the field.

In the presence of a strong saturating magnetic field, when the amplitude of the field is greater than the local magnetic field due to the dipole–dipole interaction of the impurities, the process of polarization of the nuclei changes radically. In fact, we shall derive an equation describing the nuclear polarization in the case of a strong saturating field in the low-temperature region. To simplify the subsequent calculations it is expedient to go over to an effective coordinate frame by means of the transformations

$$S_n^x\to S_n^x\cos\theta_n+S_n^z\sin\theta_n,$$

$$S_n^y\to S_n^y, \quad S_n^z\to S_n^x\sin\theta_n+S_n^z\cos\theta_n, \quad (3)$$

where

$$\cos\theta_n=\Delta_n/\Omega_{eff}, \quad \sin\theta_n=\omega_1/\Omega_{eff},$$

$$\Delta_n=\omega_S+\omega_n-\Omega, \quad \Omega_{eff}=(\Delta_n^2+\omega_1^2)^{1/2}.$$

In this coordinate frame the Hamiltonian is written in the following way:

$$\tilde{\mathcal{H}}=\sum_n \Omega_{eff}^n S_n^z-\omega_I\sum_i I_i^z+\tilde{\mathcal{H}}_d^{sec}+\tilde{\mathcal{H}}_{IS}, \tag{4}$$

where

$$\tilde{\mathcal{H}}_d^{sec}=\sum_{n,m}\lambda(\theta_{nm})[{}^1/_2 A_{nm}S_n^zS_m^z+B_{nm}S_n^+S_m^-]+{}^1/_2\sum_{n,m}\varkappa(\theta_{nm})A_{nm}S_n^zS_m^z,$$

$$\tilde{\mathcal{H}}_{IS}=\sum_{in}V_{in}^{zz}I_i^zS_n^z\cos\theta_n-{}^1/_2\sum_{in}V_{in}^{zz}I_i^z(S_n^++S_n^-)\sin\theta_n$$

$$+{}^1/_2\sum_{in}(V_{in}^{+z}I_i^-+V_{in}^{-z}I_i^+)[S_n^z\cos\theta_n-{}^1/_2(S_n^++S_n^-)\sin\theta_n],$$

$$\lambda(\theta_{nm})={}^1/_2(\cos\theta_n\cos\theta_m-2\sin\theta_n\sin\theta_m+1),$$

$$\varkappa(\theta_{nm})=1-\cos(\theta_n-\theta_m).$$

We shall assume, as is done in Refs. 6 and 7, that the strong saturating field and the spectral diffusion rapidly establish a single temperature in the electron spin system. In this case we shall not take into account the spin-lattice interaction of the impurities, for the following reasons. It is well known that the effect of the lattice is manifested in the appearance of the ratio $\alpha=T_{zL}/T_{dL}$ of the relaxation times of the Zeeman subsystem and of the dipole reservoir of the electron spins in the denominator of the expression for the stationary temperature. In our case, however, when $\Delta^*\gg\omega_d$ (Δ^* is the width of the inhomogeneously broadened EPR line), this ratio is equal to unity: $\alpha=1$. Therefore, even in conditions in which the electron spins interact with the lattice, the expression for the stationary temperature of the impurities does not contain the relaxation times.[6,7] Consequently, inclusion of the lattice does not affect the final temperature of the spins, or, by the same token, the time of establishment of the stationary polarization. Thus, after a certain short interval of time has elapsed the electron-nuclear spin system can be represented in the form of two thermodynamic subsystems: an electron subsystem with Hamiltonian

$$\mathcal{H}_S=\sum_n\Omega_{eff}^nS_n^z+\tilde{\mathcal{H}}_d^{sec}$$

and temperature β_S^{-1}, and a nuclear subsystem with Hamiltonian

$$\mathcal{H}_I=-\omega_I\sum_i I_i^z$$

and temperature β_I^{-1}.

The electron-nuclear interaction, which we have treated as a perturbation, leads to equalization of the electron and nuclear temperatures. This process can be described by the method of the nonequilibrium statistical operator,[12] by means of which it is not difficult to obtain the following equation for the inverse nuclear temperature β_I:

$$d\beta_I/dt=-(\beta_I-\beta_S)/T_1, \tag{5}$$

$$T_1^{-1}=-\frac{\pi}{8}\omega_1^2V^2\int_{-\infty}^{\infty}d\omega\,g(\omega)\frac{\varphi(\Omega_{eff}-\omega_I)}{\Omega_{eff}}\operatorname{ch}\frac{\beta_I\omega_I}{2}\operatorname{ch}^{-1}\frac{\beta_S\Omega_{eff}}{2}\frac{e^{\omega_I(\beta_S-\beta_I)}-1}{\omega_I(\beta_I-\beta_S)}$$

$$\times\exp\left\{-\frac{\beta_I\omega_I}{2}+\frac{\beta_S\Omega_{eff}}{2}\right\}-\frac{\pi}{8}V^2\int_{-\infty}^{\infty}d\omega\,g(\omega)f(\omega_I)\frac{\Delta^2}{\Omega_{eff}^2}\operatorname{ch}\frac{\beta_I\omega_I}{2}$$

$$\times\frac{[e^{-\omega_I(\beta_I-\beta_S)}-1]e^{\beta_I\omega_I/2}-[e^{\omega_I(\beta_I-\beta_S)}-1]e^{-\beta_I\omega_I/2}}{\omega_I(\beta_I-\beta_S)}, \tag{6}$$

where $f(\omega)$ and $\varphi(\omega)$ are the Fourier transforms of the following correlation functions:

$$f(t)=\frac{\operatorname{Sp}\exp\{-\beta_S\tilde{\mathcal{H}}_S\}S_n^z(t)S_n^z}{\operatorname{Sp}\exp\{-\beta_S\tilde{\mathcal{H}}_S\}(S_n^z)^2}$$

$$\varphi(t)=\frac{\operatorname{Sp}\exp\{-\beta_S\tilde{\mathcal{H}}_S\}\bar{S}_n^+(t)S_n^-}{\operatorname{Sp}\exp\{-\beta_S\tilde{\mathcal{H}}_S\}S_n^+S_n^-},$$

$$\bar{S}_n^+(t)=\exp\{i\tilde{\mathcal{H}}_dt\}S_n^+\exp\{-i\tilde{\mathcal{H}}_dt\}.$$

In the derivation of Eq. (6) we have gone over to a continuous description of the inhomogeneously broadened EPR line, i.e., we have replaced the summation over the local Zeeman energies by integration, in the following way:

$$\sum_{n=1}^{N}(\ldots)\to N\int_{-\infty}^{\infty}d\omega\,g(\omega)(\ldots),\qquad V^2=\frac{1}{N}\sum_{i,n}|V_{in}^{+z}|^2.$$

In Eq. (6) the first term describes the nuclear polarization due to the action of the alternating magnetic field and the second term determines the change in the nuclear polarization by the spin–spin interaction with the magnetic ions.

To simplify the following analysis we shall approximate $\varphi(\omega)$ by a δ-function.[3)] Then, near the stationary state, where $(\beta_S-\beta_I)\omega_I\ll1$, T^{-1} is written in the following form:

$$T_1^{-1}=\frac{\pi}{8}\frac{\omega_1^2V^2}{\omega_I(\omega_I^2-\omega_1^2)^{1/2}}[g(\omega_+)+g(\omega_-)]$$

$$+\frac{\pi}{4}V^2\operatorname{ch}^2\frac{\beta_S\omega_I}{2}\int_{-\infty}^{\infty}d\omega\,g(\omega)f(\omega_I)\frac{\Delta^2(\omega)}{\Omega_{eff}^2(\omega)}, \tag{7}$$

where $\omega_\pm=\omega_S-\Omega^\pm(\omega_I^2-\omega_1^2)^{1/2}$. Since the correlation function $f(\omega_I)$ falls off rapidly ($\omega_I\gg\omega_d$), the rate of establishment of the stationary polarization for $\omega_I\gg\omega_1$ is entirely determined by the first term.

The appearance of the singularity in (7) is connected with the approximation of $\varphi(\omega)$ by the function $\delta(\omega)$. In reality, $\varphi(\omega)$ has a width of the order of ω_d, and, therefore, the minimum value of the denominator in (7) will be of the order of ω_d, as a result of which the increase in the rate of mixing at $\omega_1=\omega_I$ will be bounded. As can be seen from the expression (7), the rate of establishment of the polarization increases strongly at $\omega_1\lesssim\omega_I$. Physically, this is connected with the fact that in the rotating coordinate frame the regular part of the energy of the electron spins is close to the nuclear frequency, and, therefore, strong exchange occurs.

When $\omega_1>\omega_I$ the first term goes to zero; therefore, the contact between the nuclear spins and the dipole reservoir of the paramagnetic impurities is very weak. The rate of establishment of the stationary nuclear polarization depends on the lattice temperature through

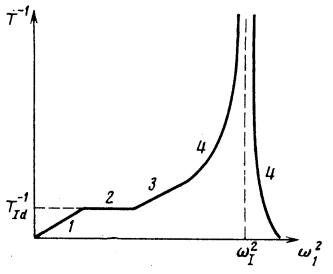

FIG. 1. Dependence of the rate of polarization on the intensity of the alternating field.

β_S. As follows from Refs. 7 and 9, as the lattice temperature is lowered β_S reaches saturation, i.e., does not change further. Therefore, the rate of establishment of the stationary nuclear polarization depends weakly on the lattice temperature. This fact was observed in the experiments of Refs. 8.

When $\omega_1 \ll \omega_I$, in the high-temperature limit, formula (7) for T^{-1} goes over into the well-known expression for the rate of nuclear polarization obtained earlier in Ref. 3.

Thus, it follows finally that:

1. If the saturating field is weak, so that $W < T_{dI}^{-1}$, the time of establishment of the stationary nuclear polarization will be proportional to the probability of transitions caused by the alternating field, i.e., will be proportional to the intensity of the alternating field. Estimates show that the experiments of Ref. 13 correspond to this case.

2. With increase in the power of the saturating field the time of establishment of the stationary polarization becomes independent of the amplitude of this field.

3. Further increase in the intensity of the alternating field can increase the probability of forbidden transitions, and then the coupling of the dipole–dipole reservoir and the nuclear system will be due to the forbidden transitions. Once again the rate of establishment of the stationary nuclear polarization will be proportional to the intensity of the alternating field. Evidently, precisely this case was realized in the experiments of Ref. 9.

4. On further increase of the amplitude of the field the dependence of the rate of establishment of the nuclear polarization on the intensity of the field increases sharply and the rate reaches a maximum as ω_1 approaches ω_I, $\omega_1 \lesssim \omega_I$. Further increase of ω_1 makes energy exchange between the dipole–dipole reservoir of the impurities and the Zeeman nuclear system more difficult, and the rate of polarization decreases sharply.

The described dependence of the rate of establishment of the stationary nuclear polarization on the intensity of the alternating field is shown schematically in Fig. 1.

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[2)] We do not take forbidden transitions into account, since their probability in this case is much smaller than the rate of the relaxation of nuclei to the dipole reservoir of the impurities that arises from their direct contact.

[3)] This is possible here, inasmuch as the EPR linewidth due to the spin-spin interaction is much smaller than the linewidth due to the causes of the inhomogeneous broadening: $\omega_d \ll \Delta^*$.

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