Cooling of a system of nuclear spins in a semiconductor upon polarization of the electron polarization

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The cooling of a nuclear spin system is considered under conditions of optical orientation, when the spin **S** of the photoelectrons and the external magnetic field oscillate at a certain frequency ω. It is shown that the singularities that arise in experiments with modulation are due to the delay of the electron-induced nonequilibrium spin relative to **S**. Expressions are obtained for the temperature of a nuclear spin system in fields on the order of the local field H_L at various values of the modulation frequency, of the phase shift, and of the angle between the electron spin and the alternating magnetic field. It is shown that experiments with modulation yield the form of the nuclear spin correlators in the region of weak magnetic fields.

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1. When the electrons of a semiconductor in a magnetic field are optically oriented, the nuclei of the crystal lattice become polarized.[1,2] This effect is a consequence of the lowering of the nuclear spin-system temperature on account of the contact with the nonequilibrium spins of the photoelectrons. As shown by D'yakonov and Perel',[3] if the electron polarization is constant in time the nuclear spin temperature is given by

$$\frac{1}{\Theta}=\frac{4I}{\mu_I}\frac{(\mathbf{HS})}{H^2+\bar{H}_L^2},\qquad (1)$$

where μ_I are the values of the magnetic moment and of the spin of the nuclei, **S** is the average spin of the photoexcited electrons, **H** is the constant magnetic field in which the cooling takes place, and H_L is a characteristic magnetic field of the order of the field produced at the nucleus by its nearest neighbors.

Fleisher *et al.*[4] investigated experimentally the cooling of a nuclear spin system with the exciting light excited at high frequency, when the nonequilibrium electron spin varied like $\mathbf{S}(t)=\mathbf{S}\cos\omega t$. It was shown that if the external magnetic field H is constant in time, there is no cooling. Tuning on a weak alternating field that oscillates at the same frequency as the spin $\mathbf{S}(\mathbf{H}_1(t)=\mathbf{H}_1\cos(\omega t+\varphi))$ leads to a noticeable cooling of the nuclear spins. The temperature Θ took on both positive and negative values, depending on the phase shift φ.

It might seem that the experimental results are a direct consequence of Eq. (1). It is actually seen from (1) that if the magnetic field is constant while **S** oscillates and $\omega T_1\gg 1$ (T_1 is the nuclear longitudinal-relaxation time) then the value of $1/\Theta$ averaged over the period of the oscillations is zero. No cooling of the nuclear spins takes place in this case. If the magnetic field likewise oscillates at the frequency ω, then

$$\frac{1}{\Theta}=\frac{2I}{\mu_I}\frac{(\mathbf{H}_1\mathbf{S})}{H^2+\bar{H}_L^2}\cos\varphi\qquad (2)$$

and the temperature of the nuclear spin is maximal in magnitude and has different signs at $\varphi=0$ and $\varphi=-\pi$.

Such reasoning, however, cannot explain the entire aggregate of the now available experimental results. Thus, Novikov and Fleĭsher[5,6] have shown that at constant electron polarization the nuclear spins can be cooled both in an external magnetic field and in the effective field $\mathbf{H}_e=h_e\mathbf{S}$ produced at the nuclei by the polarized electrons. The electron field is of the order of several oersteds and in the case of high-frequency modulation **S** oscillates in phase with the photoelectron spin ($\varphi=0$). On the basis of Eq. (2) it is readily found that **S** should be weaker by only a factor of two in the case of high-frequency modulation than at $\omega=0$. No cooling of the nuclear spin system in an electric field was observed in Ref. 4, however.

We show in the present paper that from the point of view of the cooling process there is no significant difference between the electron field and the external field, both when **S** is constant and in the case of high-frequency modulation of **S** and **H**. The singularities that arise in experiments with modulation are due to the delay of the electron-induced nonequilibrium nuclear spin **I**′ relative to **S**. Since the energy flux into the nuclear system is proportional to the relaxation rate $\dot{E}\sim(\mathbf{H}_1\mathbf{I}')$ of the nonequilibrium spin, the optimal cooling conditions are reached when **H** oscillates in phase with **I**′ rather than with **S**. At small ω, the vector **I**′ oscillates in phase with **S**, and consequently the maximum cooling is reached when the oscillations of the alternating magnetic field and of the electron spin are in phase. At $\omega T_2\gtrsim 1$, however (T_2 is the relaxatuon time of the nonequilibrium nuclear spin) the oscillations of **I**′ lag the oscillations of **S**, and maximum cooling corresponds to a nonzero phase shift φ. It is easy to satisfy this condition experimentally in the case of an external alternating magnetic field. At the same time, cooling in the electron field, which is always in phase with **S**, is much less effective in the case of high-frequency modulation than at $\omega=0$.

We note also that a radio-frequency magnetic field leads to additional relaxation of the nuclear polarization as is manifest by "heating" of the nuclear spin system.

2. In semiconductors of the GaAs type, oriented electrons polarize the lattice nuclei via contact interaction

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$$\hat{V}_{sf}=\frac{16\pi}{3I}\mu_0\mu_I\sum_{jn}\hat{\rho}_j(\mathbf{R}_n)(\hat{\mathbf{S}}_j\hat{\mathbf{I}}_n). \tag{3}$$

Here $\hat{\rho}_j(\mathbf{R})=\delta(\mathbf{r}_j-\mathbf{R})$ is the density operator of the electron numbered j at the point $\mathbf{R}$; μ_0, $\mathbf{S}_j$, $\mathbf{r}_j$ are respectively the Bohr magneton, the spin operator, and the radius vector of this electron, while μ_I, $\hat{\mathbf{I}}_n$, $\mathbf{R}_n$ are the magnetic moment, the spin operator, and the radius vector of the n-th nucleus.[1]

For further calculations it is convenient to represent $\hat{V}_{sf}$ in a somewhat different form, separating explicitly its mean value over the electron ensemble: $\hat{V}_{sf}=\langle\hat{V}_{sj}\rangle+\hat{V}$. In fact,

$$\langle\hat{V}_{sf}\rangle=-\frac{\mu_I}{I}\sum_n(\mathbf{H}_e(\mathbf{R}_n)\hat{\mathbf{I}}_n), \tag{4}$$

where

$$\mathbf{H}_e(\mathbf{R}_n)=-\frac{16\pi}{3}\mu_0\sum_j\langle\hat{\rho}_j(\mathbf{R}_n)\rangle\mathbf{S}, \tag{5}$$

$\sum_j\langle\hat{\rho}_j(\mathbf{R}_n)\rangle$ is the average electron density at the nucleus numbered n, and $\mathbf{S}$ is their average spin. The operator singled out has thus a clear-cut physical meaning—it describes the action of the effective field $\mathbf{H}_e$ of the polarized electrons (the electron field) on the nuclear spins.

The interaction of the nuclei with the fluctuations in the electron system is described by the operator $\hat{V}=\hat{V}_{sf}-\langle\hat{V}_{sf}\rangle$. This interaction makes possible the transfer of the angular momentum of the polarized electrons to the nuclear system. With (3)–(5) taken into account, the Hamiltonian of the lattice-nuclei spin system takes the form

$$\hat{\mathscr{H}}_N=-\frac{\mu_I}{I}\sum_n(\mathbf{H}(\mathbf{R}_n,t)\hat{\mathbf{I}}_n)+\hat{\mathscr{H}}_d, \tag{6}$$

where $\mathbf{H}$ is the total magnetic field acting on the lattice nuclei and is the sum of the electron and external fields, while $\hat{\mathscr{H}}_d$ is the energy operator of the spin-spin interactions of the nuclei, which we shall call for brevity dipole interactions.

The dependence of $\mathbf{H}$ on $\mathbf{R}$ is due to the fact that the density and polarization of the electrons are in general different at different lattice nuclei. The time variation of the total magnetic field can be due either to the application of an alternating magnetic field to the sample or to changes of the polarization of the electron spins. We assume here that $\mathbf{H}=\mathbf{H}_0+\mathbf{H}_1(t)$, where $\mathbf{H}_0$ is the magnetic-field component that is constant in time, and $\mathbf{H}_1(t)$ is the alternating component of $\mathbf{H}$ and oscillates at a certain frequency ω that coincides with the electronspin modulation frequency.

The behavior of the nuclear spin system density matrix Φ interacting with polarized electrons will be described by the kinetic equation[3]

$$i\hbar\frac{\partial\Phi}{\partial t}=[\hat{\mathscr{H}}_N(t),\Phi(t)]-\frac{i}{\hbar}\int_0^\infty d\tau\,\mathrm{Sp}_e[\hat{V}_\tau,[\hat{V},f(t)\Phi(t)]]. \tag{7}$$

Here $f(t)$ is the value of the density matrix at the instant of time t, Sp_e stands for the trace over the electron quantum numbers, $\hat{V}_\tau$ is given by

$$\hat{V}_\tau=\exp(i\hat{\mathscr{H}}_e\tau/\hbar)\hat{V}\exp(-i\hat{\mathscr{H}}_e\tau/\hbar), \tag{8}$$

and $\hat{\mathscr{H}}_e$ is the Hamiltonian of the electron system.

Equation (7) was derived in Ref. 3 using the fact that the values of τ that are essential in the integral term are limited by the correlation time of the orbital motion of the electrons, so that the changes that take place in the nuclear subsystem can be neglected. It was also assumed in Ref. 3 that $\hat{\mathscr{H}}_N$, $\hat{\mathscr{H}}_e$, and f are independent of time. In the case of modulation of the electron polarization and of the external magnetic field this assumption is incorrect. However, the changes of the operators $\mathscr{H}_N$, $\mathscr{H}_e$, and f during the time of the orbital correlations are small, so that (7) can be employed by introducing in it the explicit dependence of these operators on t.

We rewrite the kinetic equation, separating explicitly the average spin of the oriented electrons:

$$i\hbar\frac{\partial\Phi}{\partial t}=[\hat{\mathscr{H}}_N(t),\Phi(t)]+\sum_{\substack{mn\\ \alpha\beta\gamma}}\frac{a_{mn}}{2\hbar}\varepsilon_{\alpha\beta\gamma}S_\gamma(t)[\hat{I}_\alpha{}^n,\{\hat{I}_\beta{}^m\Phi\}]-\frac{i}{\hbar}\hat{F}(\Phi), \tag{9}$$

where $\hat{I}_\alpha{}^n$ is the operator of the component α of the spin of the nucleus numbered n, $\{\hat{I}_\beta{}^m\Phi\}=\hat{I}_\beta{}^m\Phi+\Phi\hat{I}_\beta{}^m$, $\varepsilon_{\alpha\beta\nu}$ is a unit antisymmetric tensor of third rank,

$$\hat{F}(\Phi)={}^1/_4\sum_{mn\alpha}a_{mn}[\hat{I}_\alpha{}^n,[\hat{I}_\alpha{}^m,\Phi]], \tag{10}$$

$$a_{mn}=\left(\frac{16\pi}{3I}\mu_0\mu_I\right)^2\sum_i\int_0^\infty d\tau(\langle\hat{\rho}_i(\mathbf{R}_n,0)\hat{\rho}_i(\mathbf{R}_m,\tau)\rangle-\langle\hat{\rho}_i(\mathbf{R}_n)\rangle\langle\hat{\rho}_i(\mathbf{R}_m)\rangle), \tag{11}$$

$$\langle\hat{\rho}_i(\mathbf{R}_n,0)\hat{\rho}_i(\mathbf{R}_m,\tau)\rangle=\mathrm{Sp}_e(f\hat{\rho}_i(\mathbf{R}_n,0)\hat{\rho}_i(\mathbf{R}_m,\tau)),$$

and the dependence of the operators $\hat{\rho}_i(\mathbf{R}_m,\tau)$ on τ is tiven by a formula similar to (8).[2]

On going from (7) to (9) we assumed that the correlator $\langle\hat{S}_\alpha(0)\hat{S}_\beta(\tau)\rangle$ remains practically constant for all values of τ that are of importance in the evaluation of the integral with respect to time. This assumption is valid in magnetic fields that are not too strong, when it is possible to neglect in the integral term of (7) the terms of order $\omega_e\tau$ (ω_e is the Larmor-precession frequency of the electron spin in the magnetic field).

Equation (9) describes all the principal processes that take place in a nuclear spin system optically oriented in an alternating magnetic field. The interaction of the nuclear spins with the alternating field in (9) is determined by the explicitly time-dependent terms of the Hamiltonian

$$\hat{\mathscr{H}}_N(t)=\hat{\mathscr{H}}_{N0}-\sum_n(\mathbf{H}_1(\mathbf{R}_n,t)\hat{\mathbf{I}}_n)\mu_I/I.$$

The polarization of the nuclei by oriented electrons is described by the terms that contain the time dependent average spin $\mathbf{S}(t)$. Finally, the operator $\hat{F}$ accounts for the nuclear relaxation on electrons in the conduction band.

If the magnetic field H_1 and the polarization of the electrons oscillate with time at the frequency ω, then it is convenient to resolve the sought density matrix Φ into components:

$$\Phi(t)=\sum_{n=0}^{\infty}\Phi_n(\Omega_n,t),$$

each of which oscillates at a definite frequency $\Omega_n = n\omega$. In the case when $\omega T_1 \gg 1$ the terms of this sum decrease with increasing n like $(\omega T_1)^{-n}$, so that we confine ourselves to the approximation $\Phi \approx \Phi_0 + \Phi_1$. In this case

$$i\hbar\frac{\partial\Phi_0}{\partial t}=[\hat{\mathscr{H}}_{N0},\Phi_0]+\bar{L}(t,\Phi_1)-\frac{i}{\hbar}F(\Phi_0), \tag{12}$$

$$i\hbar\frac{\partial\Phi_1}{\partial t}=[\hat{\mathscr{H}}_{N0},\Phi_1]+L(t,\Phi_0). \tag{13}$$

Here

$$L(t,\Phi)=-\left(\frac{\mu_I}{I}\right)\sum_n[(\mathbf{H}_1(\mathbf{R}_n,t)\hat{\mathbf{I}}_n),\Phi]+\sum_{\substack{mn\\ \alpha\beta\gamma}}\frac{a_{mn}}{2\hbar}\varepsilon_{\alpha\beta\gamma}S_\gamma(t)[\hat{I}_\alpha{}^n,\{\hat{I}_\beta{}^m,\Phi\}] \tag{14}$$

is an operator constituting the sum of all the explicitly time-dependent operators of the kinetic equation (9). The bar over the operator $\bar{L}$ in (12) means averaging over the modulation period $T=2\pi/\omega$.[3]

If $T_1 \gg T_2$ ($T_2 \sim 10^{-4}$ sec is the characteristic time of the spin-spin interactions), the state of the nuclear spin system can be described within the framework of the spin-temperature concept.[7] In the high temperature approximation

$$\Phi_0=1-\hat{\mathscr{H}}_{N0}\beta, \tag{15}$$

where $\beta = 1/\Theta$ is the reciprocal spin temperature of the lattice nuclei.[4] Substituting this expression in (12), multiplying the right- and left-hand sides of the obtained expression by $\hat{\mathscr{H}}_{N0}$, and calculating the trace over the quantum numbers of the nuclei, we obtain an equation for β:

$$\frac{d\beta}{dt}=\left(\frac{I}{\mu_I}\right)^2\frac{i}{\hbar}\frac{\mathrm{Sp}(\hat{\mathscr{H}}_{N0}\bar{L}(t,\Phi_1))}{\mathrm{Sp}\,I_z^2(H_0^2+H_L^2)}-\frac{\beta}{2\hbar^2N(H_0^2+H_L^2)}\sum_n a_{nn}(H_0^2+\tilde{H}_L^2), \tag{16}$$

where N is the total of nuclei, I_z is the operator of z-projection of the total spin of the lattice nuclei, H_0^2 is the square of the constant magnetic field,

$$H_L^2=\left(\frac{I}{\mu_I}\right)^2\frac{\mathrm{Sp}\,\hat{\mathscr{H}}_d^2}{\mathrm{Sp}\,I_z^2} \tag{17}$$

is the square of the local field produced at the nucleus by the neighboring nuclei, and

$$\tilde{H}_L^2=(I/\mu_I)^2N\left\{\sum_{mn\alpha}a_{mn}\,\mathrm{Sp}[\hat{\mathscr{H}}_d,\hat{I}_\alpha{}^n][\hat{I}_\alpha{}^m,\hat{\mathscr{H}}_d]\right\}\Big/\,2\,\mathrm{Sp}\,I_z^2\sum_n a_{nn}. \tag{18}$$

The second term in the right-hand side of (16) is proportional to $\mathrm{Sp}(\hat{\mathscr{H}}_{N0}F(\Phi_0))$. It corresponds to heating of the nuclear spin system via relaxation on the electrons. The first term in the right-hand side of this equation describes the rate of change of the temperature of the nuclear spin on account of interaction with the alternating field and with the polarized electrons.

Simple estimates (see the Appendix) show that the terms containing the electron spin can be neglected in the operator L. Then

$$\frac{i}{\hbar}\mathrm{Sp}(\hat{\mathscr{H}}_{N0}\bar{L}(t,\Phi_1))=-\left(\frac{\mu_I}{I}\right)\frac{1}{T}\int_0^T\sum_{n\alpha}H_{1\alpha}(\mathbf{R}_n,t)\,\mathrm{Sp}(\hat{\dot{I}}_\alpha{}^n\Phi_1)\,dt. \tag{19}$$

Here

$$\hat{\dot{I}}_\alpha{}^n=\frac{i}{\hbar}[\hat{\mathscr{H}}_{N0},\hat{I}_\alpha{}^n] \tag{20}$$

is the operator of the rate of change of the spin projection α of the nucleus numbered n on account of the interaction with the constant magnetic field and of the spin-spin interactions with the surrounding nuclei.

Equation (19) has a simple physical meaning. When a spin relaxes, its energy in the magnetic field H_1 goes over into heat. The heat flux per unit time is equal to $-(\mathbf{H}_1\mathbf{I}')\mu_I/I$, where $\mathbf{I}'$ is the rate of change of the nonequilibrium spin via interaction with the field $\mathbf{H}_0$ and via dipole interaction. The integral in the right-hand side of (19) is the mean value of this expression over the period of $\mathbf{H}_1$.

We assume that $\mathbf{S}(t)=\mathbf{S}\cos\omega t$ and that all the projections of the field $\mathbf{H}_1$ oscillate at the same frequency ω with a certain phase shift φ_α ($H_{1\alpha}(\mathbf{R}_n,t) = H_{1\alpha}(\mathbf{R}_n)\cos(\omega t+\varphi_\alpha)$). Substituting (15) in (13) and solving the equation with respect to Φ_1, we have

$$\Phi_1(t)=\mathrm{Re}\sum_{m\gamma}e^{i\omega t}\left\{H_{1\gamma}(\mathbf{R}_m)\left(\frac{\mu_I}{I}\right)\beta e^{i\varphi_\gamma}\int_{-\infty}^0\hat{\dot{I}}_\gamma{}^m(\tau)e^{i\omega\tau}d\tau+\frac{2a_{mm}}{\hbar^2}S_\gamma\int_{-\infty}^0\hat{I}_\gamma{}^m(\tau)e^{i\omega\tau}d\tau\right\}, \tag{21}$$

where

$$I_\gamma{}^m(\tau)=\exp(+i\hat{\mathscr{H}}_{N0}\tau/\hbar)I_\gamma{}^m\exp(-i\hat{\mathscr{H}}_{N0}\tau/\hbar). \tag{22}$$

Substituting this solution in (19), we get

$$\frac{i}{\hbar}\mathrm{Sp}\,\hat{\mathscr{H}}_{N0}\bar{L}(t,\Phi_1)=\frac{\mathrm{Sp}\,I_z^2}{2N}\left(\frac{\mu_I}{I}\right)\sum_{\substack{nm\\ \alpha\gamma}}H_{1\alpha}(\mathbf{R}_n)\times\left\{\frac{2a_{mm}}{\hbar^2}S_\gamma\,\mathrm{Re}\exp\{-i\varphi_\alpha\}(\delta_{nm}\delta_{\alpha\gamma}-i\omega g_{\alpha\gamma}^{nm}(\omega,\omega_0))-\omega^2\left(\frac{\mu_I}{I}\right)\beta H_{1\gamma}(\mathbf{R}_m)\,\mathrm{Re}\exp\{i(\varphi_\gamma-\varphi_\alpha)\}g_{\alpha\gamma}^{nm}(\omega,\omega_0)\right\}. \tag{23}$$

Here

$$g_{\alpha\gamma}^{nm}(\omega,\omega_0)=N\int_{-\infty}^0\frac{\langle\hat{I}_\alpha{}^n\hat{I}_\gamma{}^m(\tau)\rangle}{\mathrm{Sp}\,I_z^2}e^{i\omega\tau}d\tau \tag{24}$$

is the Fourier transform of the correlator $\langle\hat{I}_\alpha{}^n\hat{I}_\nu{}^m(\tau)\rangle = \mathrm{Sp}(\hat{I}_\alpha{}^n\hat{I}_\nu{}^m(\tau))$ and depends on the nuclear-spin Larmor-precession frequency in the constant magnetic field ($\omega_0 = \mu_I H_0/\hbar I$), and I_z^2 is the operator of the square of the z projection of the total spin of the lattice nuclei. In the derivation of (23) we used the relation

$$\int_{-\infty}^0\hat{\dot{I}}_\gamma{}^m(\tau)e^{i\omega\tau}d\tau=\hat{I}_\gamma{}^m-i\omega\int_{-\infty}^0\hat{I}_\gamma{}^m(\tau)e^{i\omega\tau}d\tau. \tag{25}$$

We note that to find Φ_1 we took into account both $\mathbf{H}_1$ and $\mathbf{S}$ in the operator $L(t,\Phi_0)$, while in the operator $\bar{L}(t,\Phi_1)$ (12) the terms containing the electron spin were left out, being small compared with the terms containing the magnetic field $\mathbf{H}_1$. The reason is that the terms with $\mathbf{H}_1$ are multiplied by β in the operator $L(t,\Phi_0)$. Since β is small, these terms can no longer be regarded as larger than the terms that contain $\mathbf{S}$.

Using (23) and (16) we obtain the final equation for the reciprocal temperature of the lattice nuclei:

$$\frac{d\beta}{dt}=G-\frac{1}{T_{1e}}\beta-\frac{1}{T_H}\beta, \tag{26}$$

where

$$G=\frac{I/\mu_I}{\hbar^2N(H^2+H_L^2)}\sum_{mn\alpha\gamma}H_{1\alpha}(\mathbf{R}_n)a_{mm}S_\gamma\operatorname{Re}\exp\{-i\varphi_\alpha\}(\delta_{nm}\delta_{\alpha\gamma}-i\omega g_{\alpha\gamma}^{nm}(\omega,\omega_0)) \tag{27}$$

is the rate of cooling of the nuclear spin system on account of the polarization of the nuclei by oriented electrons in the alternating magnetic field,

$$T_{1e}=2\hbar^2N(H_0^2+H_L^2)\Big/\sum_n a_{nn}(H_0^2+\tilde{H}_L^2) \tag{28}$$

is the characteristic time that describes the rate of relaxation of the nuclear spins on the electrons in the conduction band[3] (T_{1e} does not depend on the modulation frequency ω);

$$T_H=2N(H^2+H_L^2)/\omega^2\sum_{nm\alpha\gamma}H_{1\alpha}(\mathbf{R}_n)H_{1\gamma}(\mathbf{R}_m)\operatorname{Re}\exp\{i(\varphi_\gamma-\varphi_\alpha)\}g_{\alpha\gamma}^{nm}(\omega,\omega_0) \tag{29}$$

is the time that characterize the rate of heating of the spin system of the lattice nuclei in the alternating field $\mathbf{H}_1$. (An expression for T_H was obtained by D'yakonov and Perel'[3]). We note that Eq. (26) can contain in addition terms that describe other relaxation mechanisms (e.g., nuclear-spin relaxation on phonons).

Under stationary conditions we have $d\beta/dt=0$. The temperature of the nuclear spin system is then

$$\beta=\frac{T_{1e}T_H}{T_{1e}+T_H}G=(I/\mu_I)2\sum_{mn\alpha\gamma}H_{1\alpha}(\mathbf{R}_n)a_{mm}S_\gamma$$
$$\times\operatorname{Re}\exp\{-i\varphi_\alpha\}(\delta_{nm}\delta_{\alpha\gamma}-i\omega g_{\alpha\gamma}^{nm}(\omega,\omega_0))\Big/\Big\{\sum_n a_{nn}(H_0^2+\tilde{H}_L^2)$$
$$+(\hbar\omega)^2\sum_{\substack{nm\\ \alpha\gamma}}H_{1\alpha}(\mathbf{R}_n)H_{1\gamma}(\mathbf{R}_m)\operatorname{Re}\exp\{i(\varphi_\gamma-\varphi_\alpha)\}g_{\alpha\gamma}^{nm}(\omega,\omega_0)\Big\}. \tag{30}$$

It is seen that β is directly proportional to the rate of cooling and inversely proportional to the combined rate of the two heating processes.

We introduce certain simplifications. We assume that the density of the electrons at the lattice nuclei and the polarization of S are homogeneous over the entire crystal. In this case $\mathbf{H}_1$ and the parameter a_{nn} that characterizes the contact interaction are the same for all nuclei of the lattice ($H_{1\alpha}(\mathbf{R}_n)=H_{1\alpha}$, $a_{nn}=a$). We assume also that all the projections of $\mathbf{H}_1$ oscillate in phase with one another ($\varphi_\alpha=\varphi$). Then

$$\frac{1}{T_{1e}}=\frac{a}{2\hbar^2}\frac{H_0^2+\tilde{H}_L^2}{H_0^2+H_L^2};\quad \frac{1}{T_H}=\frac{\omega^2}{2(H_0^2+H_L^2)}\sum_\alpha H_{1\alpha}^2\operatorname{Re}g_{\alpha\alpha}(\omega,\omega_0),$$
$$G=\frac{Ia}{\mu_I\hbar^2(H_0^2+H_L^2)}\sum_{\alpha\gamma}H_{1\alpha}S_\gamma\operatorname{Re}e^{-i\varphi}(\delta_{\alpha\gamma}-i\omega g_{\alpha\gamma}(\omega,\omega_0)), \tag{31}$$
$$\beta=(I/\mu_I)2\sum_{\alpha\gamma}H_{1\alpha}S_\gamma\operatorname{Re}e^{-i\varphi}(\delta_{\alpha\gamma}-i\omega g_{\alpha\gamma}(\omega,\omega_0)/$$
$$\Big\{H_0^2+\tilde{H}_L^2+(\hbar\omega)^2a^{-1}\sum_\alpha H_{1\alpha}^2\operatorname{Re}g_{\alpha\alpha}(\omega,\omega_0)\Big\}.$$

Here

$$g_{\alpha\gamma}(\omega,\omega_0)=\int_{-\infty}^{0}\frac{\langle\hat{I}_\alpha(0)\hat{I}_\gamma(\tau)\rangle}{\operatorname{Sp}I_z^2}e^{i\omega\tau}d\tau \tag{32}$$

is the Fourier transform of the correlator, normalized to unity, of the components α and γ of the total spin I of the lattice nuclei. In (31) we have assumed that the z axis is directed along $\mathbf{H}_0$. From symmetry consideration it follows in this case that

$$g_{xz}=g_{yz}=g_{zx}=g_{zy}=0,\quad g_{xx}=g_{yy},\quad g_{xy}=-g_{yx},$$
$$\sum_{\alpha\gamma}H_{1\alpha}H_{1\gamma}g_{\alpha\gamma}=\sum_\alpha H_{1\alpha}^2g_{\alpha\alpha}.$$

3. To investigate the dependence of the nuclear temperature on the modulation frequency, on the phase shift, and on the angle between S and $\mathbf{H}_1$ it is necessary to know the explicit forms of the correlators $\langle\hat{I}_\alpha(0)\hat{I}_\gamma(\tau)\rangle$ or of their Fourier transforms $g_{\alpha\gamma}$. In the theory of nuclear magnetic resonance these correlators are determined by using various approximations (see, e.g., Ref. 7), with all the calculations made usually for strong fields ($H_0\gg H_L$), whereas Eqs. (28)–(31) are valid at $H_0\lesssim H_L$. The analysis that follows we assume that $\langle\hat{I}_\alpha(0)\hat{I}_\gamma(\tau)\rangle$ decreases exponentially with increasing τ, and in the conclusion we shall discuss the differences between the obtained answers and the results of numerical calculations for a Gaussian approximation of the correlators.

Assume that the nonzero correlators satisfy the relations

$$\langle\hat{I}_z(0)\hat{I}_z(\tau)\rangle=\exp(-|\tau|/T_{2\parallel})\operatorname{Sp}I_z^2,$$
$$\langle\hat{I}_x(0)\hat{I}_x(\tau)\rangle=\langle\hat{I}_y(0)\hat{I}_y(\tau)\rangle=\operatorname{Sp}I_z^2\cos\omega_0\tau\exp(-|\tau|/T_{2\perp}), \tag{33}$$
$$\langle\hat{I}_y(0)\hat{I}_x(\tau)\rangle=-\langle\hat{I}_x(0)\hat{I}_y(\tau)\rangle=\operatorname{Sp}I_z^2\sin\omega_0\tau\exp(-|\tau|/T_{2\perp}),$$

where ω_0 is the Larmor precession frequency of the nuclear signals in the constant magnetic field H_0, while $T_{2\parallel}$ and $T_{2\perp}$ are the relaxation times of the longitudinal and transverse components of the nuclear spin and depend in the general case on H_0($T_{2\parallel}=T_{2\perp}$ at $H=0$). Then

$$g_{zz}=T_{2\parallel}g(\omega T_{2\parallel}),\quad g_{xx}=g_{yy}=T_{2\perp}[g((\omega+\omega_0)T_{2\perp})+g((\omega-\omega_0)T_{2\perp})]/2,$$
$$g_{xy}=-g_{yx}=iT_{2\perp}[g((\omega+\omega_0)T_{2\perp})-g((\omega-\omega_0)T_{2\perp})]/2, \tag{34}$$
$$g(x)=\int_{-\infty}^{0}e^{(1+ix)\tau}d\tau=\frac{1-ix}{1+x^2}.$$

Substituting (34) in (31) we find that if the alternating field $\mathbf{H}_1$ is directed along the magnetic field $\mathbf{H}_0$, then

$$T_H=2T_{2\parallel}\frac{(H_0^2+H_L^2)(1+x^2)}{H_1^2x^2},$$
$$G=\frac{Ia}{\mu_I\hbar^2}\frac{(\mathbf{H}_1\mathbf{S})(\cos\varphi-x\sin\varphi)}{(H_0^2+H_L^2)(1+x^2)}, \tag{35}$$
$$\beta=\frac{I}{\mu_I}\frac{2(\mathbf{H}_1\mathbf{S})(\cos\varphi-x\sin\varphi)}{(1+x^2)(H_0^2+\tilde{H}_L^2)+\xi x^2H_1^2},$$

where $x=\omega T_{2\parallel}$, and the parameter $\xi=\hbar^2(aT_{2\parallel})^{-1}\approx T_{1e}/T_{2\parallel}\gg1$. The same formula describes in the case $H_0=0$ the nuclear temperature at arbitrary directions of S and $\mathbf{H}_1$.

As seen from (35), there are three characteristic ranges of the modulation frequency. At small ω, when $x^2\xi\ll1$, Eq. (35) coincides with formula (2) obtained from the usual expression (1) for the nuclear spin temperature by substituting the oscillating S and $\mathbf{H}_1$, followed by averaging over the modulation period. In this case the delay of the nonequilibrium nuclear spin relative to S is small (the maximum cooling is reached at $\varphi=0$), and the heating in the alternating field H_1 is negligible compared with the relaxation of the nuclear spins on the electrons ($T_{1e}\ll T_H$).

If $x \ll 1$ but $x^2\xi \gg 1$, then at $H_1^2 x^2 \xi \geq H_0^2 + \tilde{H}_L^2$ the value of β can decrease noticeably on account of heating in the RF field ($T_{1e} \gtrsim T_H$). The delay of the nonequilibrium spin is here likewise small and maximum cooling corresponds to $\varphi = 0$.

Finally, at high modulation frequencies, when $\omega T_{2\parallel} \gg 1$, the minimum temperature of the nuclear spin system corresponds to a phase shift $\varphi = -\pi/2$ (the nonequilibrium spin lags S by one-quarter of a period). There is then practically no cooling in the electron field, for which $\varphi = 0$, whereas the cooling in the external field can become appreciable. A similar effect was observed in Ref. 4.

Let now $\mathbf{H}_1$ and $\mathbf{S}$ be perpendicular to $\mathbf{H}_0$. Then

$$T_H = 2T_{2\perp}\frac{(H_0^2+H_L^2)[(1+x_0^2-x^2)^2+4x^2]}{H_1^2x^2(1+x_0^2+x^2)},$$
$$G = \frac{Ia}{\mu_I\hbar^2}\frac{H_1S}{H_0^2+H_L^2}\operatorname{Re} e^{-i\varphi}\frac{\cos\alpha(1+x_0^2+ix)+ixx_0\sin\alpha}{(1+ix)^2+x_0^2}, \tag{36}$$
$$\beta = 2\frac{I}{\mu_I}H_1S\operatorname{Re} e^{-i\varphi}\frac{\cos\alpha(1+x_0^2+ix)+ixx_0\sin\alpha}{(1+ix)^2+x_0^2} \times\left(H_0^2+\tilde{H}_L^2+H_1^2\xi x^2\operatorname{Re}\frac{1+ix}{(1+ix)^2+x_0^2}\right)^{-1},$$

where $x = \omega T_{2\perp}$, $x_0 = \omega_0 T_{2\perp}$, $\xi = \hbar^2(aT_{2\perp})^{-1}$, and α is the angle between $\mathbf{S}$ and $\mathbf{H}_1$. It is seen that in this case the cooling of the nuclear spins can take place even if the scalar product $(\mathbf{S}\cdot\mathbf{H}_1) = 0$ ($\alpha = \pm\pi/2$). Thus, at small values of the field H_0, when $\omega_0 T_{2\perp} \gg 1$ and $\alpha = \pi/2$, we have for β:

$$\beta = \frac{I}{\mu_I}\frac{2H_1Sxx_0(2x\cos\varphi+(1-x^2)\sin\varphi)}{(1+x^2)[(1+x^2)(H_0^2+\tilde{H}_L^2)+H_1^2\xi x^2]}. \tag{37}$$

If $x \ll 1$, then the maximum cooling is reached at $\varphi = \pi/2$, while for $x = 1$ the value of β is a minimum at $\varphi = 0$. If, however, the modulation frequency is high, the minimum temperature corresponds to $\varphi = -\pi/2$.

An investigation of formula (36) at $\omega_0 T_2 \gg 1$ (the case of strong magnetic fields) is meaningless since, as already noted above, the nuclear spin system is described in this case not by one but by two temperatures pertaining to the Zeeman and dipole pools. As shown for example in Ref. 7, heating in an RF field (nuclear magnetic resonance) at $H_0 \gg H_L$ is less effective than given by formulas (26), (29), (31), and (36).

The foregoing principal qualitative results are independent of the concrete type of the nuclear spin correlators. These results include: 1) the possibility of cooling a nuclear spin system when all three vectors $\mathbf{S}$, $\mathbf{H}_0$, and $\mathbf{H}_1$ are perpendicular to one another, 2) the statement that at a finite modulation frequency maximum cooling corresponds to a nonzero phase shift. However, the quantitative relations for the cooling rate (G), for the heating rate in the alternating field (T_H^{-1}), and consequently also for the stationary value of the reciprocal temperature (β) as functions of ω, ω_0, φ, and α differ for different types of correlators $\langle I_\alpha(0)I_\gamma(\tau)\rangle$.

Curves 1 and 2 of Fig. 1 are plots of T_H^{-1} against the modulation frequeny ω for two values of the constant

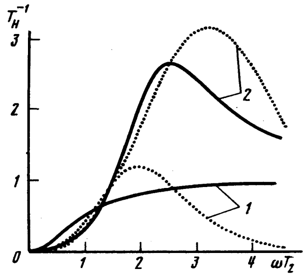

FIG. 1. Nuclear-spin relaxation rates in an alternating field for exponentially decreasing (solid curves) and Gaussian (dotted) correlators: $1-\omega_0T_2=0$, $2-\omega_0T_2=2$. The time T_H is measured in units of $2T_2(H_0^2+H_L^2)H_1^{-2}$.

magnetic field H_0($\omega_0 T_2 = 0$, $\omega_0 T_2 = 2$) in the case when $\langle I_\alpha(0)I_\nu(\tau)\rangle \sim \exp\{-|\tau|/T_2\}$ (solid curves) and $\langle I_\alpha(0)I_\gamma(\tau)\rangle \sim \exp(-(\tau/T_2)^2)$ (dotted). The alternating field $\mathbf{H}_1$ is assumed perpendicular to $\mathbf{H}_0$. It is seen that for an exponentially decreasing correlator, at high modulation frequencies, the time T_H tends to a finite value $T_H^{-1} = (2T_2)^{-1}H_1^2/(H_0^2+H_L^2)$, whereas if the fall-off of $\langle I_\alpha(0)I_\gamma(\tau)\rangle$ is Gaussian this rate tends to zero as $\omega T_2 \to \infty$. This is most clearly demonstrated by the curves calculated at $H_0 = 0$. In the case of a Gaussian correlator curve 1 a maximum has at $\omega T_2 = 2$. At $\omega T_2 > 2$ the heating rate begins to decrease with increasing modulation frequency. At the same time the curve corresponding to exponential damping increases monotonically over the entire range of values of ω.

Figure 2 shows a comparison of the nuclear-spin cooling rates for two types of correlators for the cases $\alpha = \varphi = 0$; $\alpha = 0$, $\varphi = -\pi/2$; $\omega_0 T_2 = 0$, and $\omega_0 T_2 = 2$. It is seen that at low modulation frequencies and $\varphi = -\pi/2$ there is no cooling regardless of the form of the correlator, and at $\varphi = 0$ the cooling rate is a maximum. At not too high values of ω the cooling is stronger in the case $\varphi = -\pi/2$ than at $\varphi = 0$. A substantial difference between the exponential and Gaussian correlator fall-off rates sets in at high modulation frequencies. The value $\varphi = 0$ is more effective here for the Gaussian law, and $\varphi = -/2$ for the exponential fall-off. We note that in both cases the sign of G can change at high frequencies if $\alpha = \varphi = 0$.

Figure 3 shows a plot of the reciprocal temperature

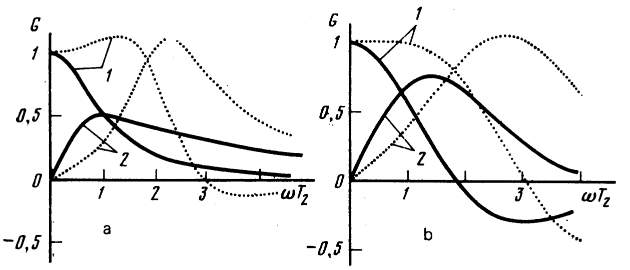

FIG. 2. Cooling rate of nuclear spin system vs. modulation frequency of oriented electron and the alternating magnetic field, for exponential (a) and Gaussian (b) correlators: $1-\alpha=\varphi=0$, $2-\varphi=-\pi/2$, $\alpha=0$. The solid lines are the results of the calculations for $\omega_0=0$, and the dotted for $\omega_0T_2=2$. The cooling rate G is measured in units of $IaH_1S(\mu_I\hbar^2)^{-1}(H_0^2+H_L^2)^{-1}$.

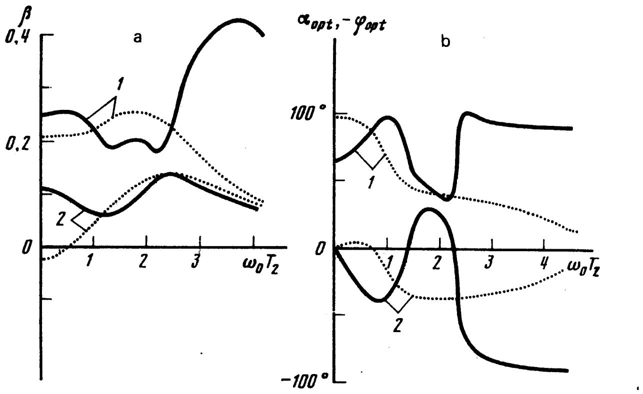

FIG. 3. Comparison of the stationary values of the nuclear spin system temperature (a) for $\alpha = \varphi = 0$ (curves 1) and for theopimal values of the phase shift and of the angle between $\mathbf{S}$ and $\mathbf{H}_1$ (curves 1). The values of $\alpha_{\rm opt}$ and $\varphi_{\rm opt}$ are shown in Fig. 3b. The solid lines correspond to exponential damping and the dotted to a Gaussian fall-off of the spin correlators with time. Curves 1 of Fig. 3b show the optimal phase shift $\varphi_{\rm opt}$ taken with a minus sign, and curves 2 show the angle $\alpha_{\rm opt}$. The calculations were made for the following parameter values: $H_1/\tilde{H}_L = 0.1$, $\omega T_2 = 2$, $\xi = 100$, $H_0/\tilde{H}_L = \omega_0 T_2$. The reciprocal temperature β is measured in units of $(I/\mu_I) 2 H_1 S \tilde{H}_L^{-2}$.

against the external magnetic field for $\alpha = \varphi = 0$ and for the optimal values of the phase shift and of the angle between $\mathbf{S}$ and $\mathbf{H}_1$. It is seen that by varying φ and α it is possible to make the nuclear spin system temperature lower by one order of magnitude than at $\alpha = \varphi = 0$.

It follows from the foregoing that modulation of the electron spin and of the magnetic field leads to qualitative changes in the conditions of the cooling of a nuclear spin system. Depending on the modulation frequency and on the dc component of the magnetic field, maximum cooling sets in at different nonzero values of the phase shift and of the angle between the oscillating $\mathbf{S}$ and $\mathbf{H}_1$. In an electron field for which $\alpha = \varphi = 0$ there may be practically no cooling. The oscillations of the magnetic field cause additional relaxation of the nuclear polarization. Experiments on the cooling of the spin system of the lattive nuclei by photo-oriented electrons whose polarization is modulated in time can help determine the form of the nuclear-spin correlator in the region of weak magnetic fields ($H_0 \lesssim H_L$).

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APPENDIX

Assume for the sake of argument that the alternating magnetic field is the effective field of the polarized electrons, $\mathbf{H}_e h_e = \mathbf{S}$. Then those terms of the operator $\overline{L}(t, \Phi_1)$ which contain $\mathbf{H}_1$ are of the order of $\mu_I h_e S$, and the terms that describe the polarization of the nuclei via contact interaction with the polarized electrons are of the order of $(\mu_I h_e)^2 S\tau'/\hbar$. Here τ' is the characteristic lifetime of the orbital correlation. This time, naturally, does not exceed the lifetime of the photoelectron, i.e., $\tau' \ll 10^{-7}$ sec. Noting that $h_e \lesssim 100$ Oe and $\mu_I \sim 10^{-23}$ erg/G, we obtain $\mu_I h_e S\{(\mu_I h_e)^2 S\tau'/\hbar\}^{-1} \approx \hbar/(\mu_I h_e \tau') \gg 10$. The terms containing the alternating magnetic field are thus larger by several orders of magnitude than the terms that describe the polarization of the nuclei by the electrons.

[1)] Here and elsewhere we assume for simplicity that all lattice nuclei are identical.

[2)] The coefficients a_{mn} can depend on t. For simplicity, we neglect this dependence hereafter.

[3)] A small term $F(\Phi_1)$ is neglected in Eq. (13).

[4)] We assume here tacitly that the constant external magnetic field H_0 is not too large compared with the random local field H_L. In the opposite case of strong external fields, the state of the nuclear spin-spin system must be described with the aid of two temperatures pertaining to the Zeeman and dipole pools.[7]

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