

Cooling of a system of nuclear spins following optical orientation of electrons in semiconductors

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A general expression is derived for the spin temperature of semiconductor lattice nuclei interacting with electrons. The expression is valid for an arbitrary disequilibrium of the electron state relative to the spin and for an arbitrary value of the external magnetic field strength. Interaction with optically oriented electrons results in a strong lowering of the nuclear spin temperature. The greatest cooling should occur when the intensity of the external magnetic field is of the same order of magnitude as that of the local field produced by dipole-dipole interaction of nuclei. It is shown that a considerable polarization of nuclei by optically oriented electrons should also be possible in a weak magnetic field.

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1. When electrons are optically oriented in semiconductors,^[1] an appreciable polarization of the crystal-lattice nuclei takes place.^[2,3] The onset of this polarization is in essence the result of cooling of the nuclear spin system, due to its interaction with the nonequilibrium electrons. Indeed, any nuclear-spin state relaxes to a thermodynamic-equilibrium state characterized by a certain nuclear spin temperature Θ within a time on the order of the time of transverse relaxation T_2 . This time is quite short in comparison with the characteristic times T_1 and T_{1e} of the change of Θ due to spin-lattice relaxation and due to interaction with the oriented electrons. Therefore during the course of the optical orientation the state of the nuclear spin system can be regarded, with good approximation, as being in thermodynamic equilibrium (the deviation from equilibrium is of the order of T_2/T_{1e}). The entire action of the photo-excited electrons reduces thus to a change of the spin temperature.

This approach enables us to extend the theory of nuclear polarization by optically oriented electrons^[3] to the region of weak external magnetic fields $H \lesssim H_L$, where H_L is the local field due to the dipole-dipole interaction of the nuclear spins. It may appear at first glance that at $H \lesssim H_L$ the nuclear polarization is proportional to the ratio T_2/T_{1e} (where T_{1e} is the time of polarization of the nuclei by the electrons, typical values of T_2/T_{1e} being of the order of $10^{-5} - 10^{-7}$), inasmuch as in weak field the transverse relaxation time T_2 characterizes simultaneously the rate at which the nonequilibrium polarization of the nuclei vanishes as a result of the transition of the Zeeman energy into the energy of the dipole-dipole interaction. In fact, this reasoning is incorrect, since it pertains only to the nonequilibrium part of the nuclear polarization. It will be shown below that oriented electrons lower the temperature of the nuclear spin system in such a way that the equilibrium nuclear polarization corresponding to this temperature and to the external magnetic field is much higher than the nonequilibrium polarization. Only in a very weak field $H < H_L(T_2/T_{1e})^{1/2}$ does the equilibrium polarization become smaller than the nonequilibrium one.

In this paper we obtain a general expression (formula (11)) for the spin temperature Θ of a nuclear system interacting with electrons; this formula is valid for an arbitrary nonequilibrium electron spin state and for an arbitrary external magnetic field. We use here

the high-temperature approximation, i.e., we assume the nuclear spin energy in the external and local fields to be small in comparison with Θ . The results of this paper show that in the case of optical orientation of the electrons the nuclear spin system can be cooled to very low temperatures, both positive or negative. The most effective cooling takes place at $H \sim H_L$, when a nuclear spin temperature $\Theta \sim \mu_I H_L$, i.e., of the order of the characteristic energy of the dipole-dipole interaction, can be attained.^[1]

2. We start with the kinetic equation for the nuclear spin density matrix Φ

$$i\hbar \frac{\partial \Phi}{\partial t} = [\mathcal{H}_N, \Phi] - \frac{i}{\hbar} \int d\tau S p_e [V_\tau, [V, f\Phi(t)]], \quad (1)$$

where

$$\mathcal{H}_N = \frac{\mu_I H}{\hbar} \sum_n I_n + \mathcal{H}_d, \quad (2)$$

where \mathcal{H}_N is the Hamiltonian of the nuclear spin system, \mathcal{H}_d is the energy operator of the dipole-dipole interaction of the nuclear spins, and μ_I and I are the magnetic moment and the spin of the nucleus. The summation in (2) is over all the lattice nuclei (we consider for the time being a simple lattice consisting of identical nuclei). In formula (1), V is the operator of the hyperfine interaction of the nuclei with the electrons

$$V = \frac{16\pi}{3I} \mu_0 \mu_I \sum_n \delta(\mathbf{r}_j - \mathbf{R}_n) S_j I_n, \quad (3)$$

where μ_0 , S_j , and \mathbf{r}_j are the Bohr magneton, the spin operator, and the radius vector of the j -th electron, while \mathbf{R}_n is the position of the n -th nucleus. The operator V_τ is given by

$$V_\tau = \exp(i\mathcal{H}_e \tau/\hbar) V \exp(-i\mathcal{H}_e \tau/\hbar),$$

where \mathcal{H}_e is the Hamiltonian of the electron subsystem. The symbol $S p_e$ in (1) denotes the trace over the electron quantum numbers, and f is the density matrix of the electron subsystem.

Equation (1) is in fact contained in Abragam's book.^[4] It was derived by recognizing that the values of τ that play an important role in the integral term of (1) are bounded by the correlation time of the orbital motion of the electron. The changes that occur during that time in the nuclear subsystem can be neglected.

As already mentioned, the nuclear spin density matrix Φ can be assumed to be in equilibrium with a

high degree of accuracy (we neglect corrections of order T_2/T_{1e}). In the high temperature approximation we have $\Phi = 1 - \mathcal{N}_N/\Theta$. Using this approximation, multiplying both halves of (1) by \mathcal{N}_N , and taking the trace over the spin quantum numbers of the nuclei, we obtain the energy balance equation for the nuclear spin subsystem, in the form

$$\frac{d}{dt} \left(\frac{1}{\Theta} \right) = - \frac{1}{T_{1e}} \left(\frac{1}{\Theta} - \frac{1}{\Theta_0} \right). \quad (4)$$

The time of the longitudinal nuclear spin relaxation on the electrons is determined here by the usual formula [4]

$$\frac{1}{T_{1e}} = \left(\frac{16\pi\mu_0\mu_I}{3\hbar} \right)^2 (Q_{xx} + Q_{yy}) \frac{H^2 + H_L^2}{H^2 + H_L^2}, \quad (5)$$

$$H_L^2 = 3I\mathcal{H}_L^2/N(I+1)\mu_I^2, \quad (6)$$

where H_L^2 is the mean squared local magnetic field produced by the surrounding nuclei at the nucleus, and

$$H_L^2 = 2H^2 \frac{Q_{xx} + Q_{yy} + Q_{zz}}{Q_{xx} + Q_{yy}}. \quad (7)$$

The state of the electron subsystem enters in (5) and (7) via the correlators $Q_{\alpha\beta}$ of the electron spin density at the nucleus

$$Q_{\alpha\beta} = \int d\tau \text{Sp}_e \{ [s_\alpha(\mathbf{R}, \tau) s_\beta(\mathbf{R}, 0) + s_\alpha(\mathbf{R}, 0) s_\beta(\mathbf{R}, \tau)] \}, \quad (8)$$

where $s_\alpha(\mathbf{R}, \tau)$ is the operator of the α -projection of the electron spin density at the nucleus at the instant τ . The z axis is chosen along the external magnetic field. Formulas (5)–(7) are valid under the assumption that the correlation radius of the electron spin density greatly exceeds the lattice constant, as is the case for semiconductors.

At $\mu_0H \ll T$, where T is the lattice temperature, we have $Q_{xx} = Q_{yy} = Q_{zz}$, and formulas (5)–(7) go over into the corresponding expressions given in Abragam's book. In particular, $H_L^2 = 3H^2$ in this case. The nuclear spin relaxation in a quantizing magnetic field was considered by a number of workers. [5] We are interested here principally in the temperature Θ_0 that becomes established under stationary conditions when the nuclear system interacts with electrons that are not in equilibrium with respect to spin. The expression obtained for Θ_0 in the derivation of (4) by the method described above is of the form

$$\frac{1}{\Theta_0} = \frac{2I}{\mu_I} \frac{H}{H^2 + H_L^2} \frac{i(Q_{yz} - Q_{zy})}{Q_{xx} + Q_{yy}} \quad (9)$$

3. The last factor in the right-hand side of (9) is expressed in universal form via the difference $\Delta\zeta = \zeta_+ - \zeta_-$ of the chemical potentials of the electrons with spins parallel and antiparallel to the external field:

$$\frac{i(Q_{yz} - Q_{zy})}{Q_{xx} + Q_{yy}} = \text{th} \frac{\Delta\zeta}{2T} \quad (10)$$

The derivation of (10) is given in the Appendix. In order for it to be valid it is necessary only that the equilibrium in a system of electrons with fixed spin projection become established much more rapidly than the spin relaxation. In other words, it is necessary that the chemical potentials ζ_+ and ζ_- have a meaning.

We thus obtain for Θ_0 the following expression, which is the main result of this paper:

$$\frac{1}{\Theta_0} = \frac{2I}{\mu_I} \frac{H}{H^2 + H_L^2} \text{th} \frac{\Delta\zeta}{2T} \quad (11)$$

The average projection of the nuclear spin on the magnetic-field direction in the stationary state is determined in terms of $\bar{I}_z = \mu_I H(I+1)/3\Theta_0$. Therefore

$$\bar{I}_z = \frac{2}{3} I(I+1) \frac{H^2}{H^2 + H_L^2} \text{th} \frac{\Delta\zeta}{2T}. \quad (12)$$

Formulas (11) and (12) do not take into account the possible "leakage" of the nuclear polarization via other relaxation mechanisms. Allowance for these mechanisms would lead to the appearance of the usual leakage factor in the right-hand sides of these formulas. We recall that our entire derivation is valid only if $\bar{I}_z \ll I$, since we used the high-temperature expansion of the nuclear spin density matrix. At $\Delta\zeta = 0$, i.e., at thermal equilibrium, it follows from (11) that $\Theta_0 \rightarrow \infty$. The reason is that in the derivation of (4) we have in fact neglected the energy of the nucleus in the magnetic field in comparison with T . In fact, as $\Delta\zeta \rightarrow 0$ the value of Θ_0 should, of course, tend to the lattice temperature T .

The quantity \tilde{H}_L^2 defined by (7) plays an important role in (11) only for a weak external field $H \lesssim H_L$. We then have $\mu_0H \ll T$ and we can assume that \tilde{H}_L does not depend on the external field. Thus, formula (11) yields the explicit dependence of Θ_0 on the external field at a fixed value of $\Delta\zeta$. As seen from this formula, the minimal value of the temperature is reached at $H = \tilde{H}_L$. Here

$$\Theta_{0\min} = \frac{\mu_I H_L}{I} \text{cth} \frac{\Delta\zeta}{2T}. \quad (13)$$

At $\Delta\zeta \gg T$ it follows from (13) that $\Theta_{\min} = \mu_I H_L/I$, but strictly speaking formula (13) does not hold in this case, since the conditions of the high-temperature approximation are violated. We can nevertheless expect $\Theta_{\min} \sim \mu_I H_L$ at $\Delta\zeta \gg T$.

4. The quantity $\Delta\zeta$ in (11) can be connected with the mean value of the electron spin projection S_Z on the magnetic-field direction. In particular, for nondegenerate electrons we have

$$n_+/n_- = \exp[(\Delta\zeta - \mu_0 gH)/T], \quad \bar{S}_z = 1/2(n_+ - n_-)/(n_+ + n_-),$$

whence

$$\text{th} \frac{\Delta\zeta}{2T} = \frac{2(\bar{S}_z - S_T)}{1 - 4\bar{S}_z S_T},$$

where $S_T = -(1/2)\tanh(\mu_0 gH/2T)$ is the equilibrium value of the electron spin, and g is the electron g -factor in the semiconductor. Thus, for nondegenerate electrons formula (11) takes the form

$$\frac{1}{\Theta_0} = \frac{4I}{\mu_I} \frac{H}{H^2 + H_L^2} \frac{\bar{S}_z - S_T}{1 - 4\bar{S}_z S_T} \quad (14)$$

Of particular interest is the case when the electron spins are fully disordered ($\bar{S}_z = 0$), in spite of the presence of an external magnetic field, as is the case in the Overhauser effect. Then, regardless of the degree of electron degeneracy, we have $\Delta\zeta = \mu_0 gH$, and formula (12) yields for the nuclear polarization

$$\bar{I}_z = \frac{2}{3} I(I+1) \frac{H^2}{H^2 + H_L^2} \text{th} \left(\frac{\mu_0 gH}{2T} \right). \quad (15)$$

This expression (so long as $\bar{I}_z \ll I$) describes the Overhauser effect in arbitrary magnetic fields, including at $H \lesssim H_L$.

5. We have considered so far a simple lattice consisting of identical nuclei. If nuclei of several sorts are present then, generally speaking the corresponding spin temperatures can be different. In weak fields

$H \sim H_L$, however, a spin temperature which is the same for all the sorts of the nuclei is rapidly established.^[4] In this case the structure of (11) remains the same as before, with the substitutions

$$\begin{aligned} \frac{2I}{\mu_I} &\rightarrow \frac{2}{\sigma} \sum_n \mu_{In} (I_n + 1) A_n^2, \\ \sigma &= \sum_n \frac{\mu_{In}^2 (I_n + 1)}{I_n} A_n^2, \\ H_L^2 &\rightarrow -\frac{3}{2\hbar^2 \sigma} \sum_{nm} \text{Sp} [I_n \mathcal{H}_d] [I_m \mathcal{H}_d] A_n A_m, \end{aligned} \quad (16)$$

where the summation is over all the crystal nuclei and

$$A_n = \frac{16\pi}{3I_n} \mu_0 \mu_{In} |u(\mathbf{R}_n)|^2 \quad (17)$$

is the constant of the hyperfine interaction of the n -th nucleus with the electrons, while $u(\mathbf{R}_n)$ is the Bloch amplitude of the wave function of the electron at the n -th nucleus.

6. We note in conclusion that the nuclear spin density matrix has in addition to its equilibrium part also a small nonequilibrium addition of order $T_2/T_1 e$. The nuclear polarization due to this addition is also proportional to $T_2/T_1 e$. The equilibrium polarization, however, according to (12), is proportional to the ratio H^2/\tilde{H}_L^2 and can become smaller than the non-equilibrium polarization in very weak magnetic fields. Thus, formula (12) for the average magnetization of the nuclei remains valid so long as $H > \tilde{H}_L (T_2/T_1 e)^{1/2}$. One can expect in weaker magnetic fields the nuclear magnetization to be determined by the nonequilibrium part of the density matrix and to be independent of the external magnetic field.

APPENDIX

DERIVATION OF FORMULA (10)

We express the Cartesian components of the tensor $Q_{\alpha\beta}$ in terms of its circular components

$$i(Q_{yx} - Q_{xy}) = Q_{--} - Q_{+-}, \quad Q_{xx} + Q_{yy} = -Q_{--} - Q_{+-}. \quad (A.1)$$

In the representation in which the electron Hamiltonian \mathcal{H}_e is diagonal we have, in accord with (8),

$$\begin{aligned} Q_{+-} &= 2\pi \sum_{ab} f_a |s_+(\mathbf{R}_n)_{ab}|^2 \delta(E_a - E_b), \\ Q_{-+} &= 2\pi \sum_{ab} f_b |s_+(\mathbf{R}_n)_{ab}|^2 \delta(E_a - E_b). \end{aligned} \quad (A.2)$$

Neglecting the spin-orbit interaction, we express the diagonal elements of the electron density matrix in the form

$$f_a = Z^{-1} \exp [-(E_a - \zeta_+ n_{+a} - \zeta_- n_{-a})/T], \quad (A.3)$$

where $n_{\pm a}$ are the numbers of electrons with spins parallel and antiparallel to the field in the state a . The matrix elements s_{+ab} connect states a and b such that $n_{+a} = n_{+b} + 1$ and $n_{-a} = n_{-b} - 1$. Recognizing that $E_a = E_b$ in (A.2), we obtain $Q_{+-}/Q_{-+} = f_a/f_b = e^{\Delta\zeta/T}$, where $\Delta\zeta = \zeta_+ - \zeta_-$. Hence, using (A.1), we obtain formula (10) of the main text.

¹⁾This qualitative conclusion can be drawn even in the framework of the high-temperature approximation, although strictly speaking it is valid only if $\Theta \gg \mu|H_L$.

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