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

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A general expression is given for the spin temperature of semiconductor lattice nuclei interacting with electrons. The expression is valid for an arbitrary disquisition of the electron system 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 perturbation due to the dipole orientation of nuclei. It is shown that a considerable polarization of nuclei by optically oriented electrons should also be possible in a weak magnetic field.

PACS numbers: 71.70.Pp

1. When electrons are optically oriented in semiconductors, an appreciable polarization of the crystal-lattice nuclei takes place. 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 nuclear spin temperature $T_N$ within a time on the order of the time of transverse relaxation $T_\perp$. This time is quite short in comparison with the characteristic times $T_\perp$ and $T_{1\rho}$ of the change of $\rho$ 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 equilibrated magnetically. The equilibrium deviation from equilibrium is of the order of $T_\perp$. 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 to the region of nonequilibrium magnetic fields $H \neq H_0$, where $H_0$ is the local field due to the dipole-dipole interaction of the nuclear spins. It may appear at first glance that in $H \neq H_0$ the nuclear polarization is proportional to the ratio $T_\perp/T_1$ (where $T_1$ is the time of polarization of the nuclear by the electrons, typical values of $T_\perp/T_1$ being of the order of $10^{-7}$ to $10^{-9}$), inasmuch as a weak field in the transverse relaxation time $T_\perp$ is expected. However, the rate at which the nonequilibrium polarization of the nuclear vanishes as a result of this relaxation is much faster than the energy into the energy of the dipole-dipole interaction. In fact, this reasoning is incorrect, since it pertains only to the equilibrium 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 polarization corresponding to this temperature and to the external magnetic field is much higher than the nonequilibrium polarization. Only in a very weak field $H_0$ does the equilibrium polarization become smaller than the nonequilibrium one.

In this paper we obtain a general expression (formula (31)) for the spin temperature of a nuclear system interacting with electrons; this formula is valid for arbitrary disquisition of nonequilibrium 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 $\Theta$. 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 $H = H_0$, when a nuclear-spin state relaxes with $\sim \mu_2H_0$, i.e., of the order of the characteristic energy of the dipole-dipole interaction, can be attained.

2. We start with the kinetic equation for the nuclear spin density matrix $\hat{\rho}$

$$\frac{\partial \hat{\rho}}{\partial t} = i[H, \hat{\rho}] - \frac{1}{T_1}[H, \hat{\rho}] - \frac{1}{T_2}[H, \hat{\rho}]$$

where $\hat{\rho}$ is the Hamiltonian of the nuclear spin system, $\hat{H}$ is the energy operator of the dipole-dipole interaction of the nuclear spins, and $\mu_1$ and $\mu_2$ are the magnetic moment and the magnetic dipole moment of the nucleus. The sum $S$ (in $\mu_1$) is over all the lattice nuclei (we consider for the time being a simple lattice consisting of identical nuclei). $\mu_2$ is the operator of the hyperfine interaction of the nuclei with the electrons.

$$\langle \hat{\rho} \rangle = \sum_{\mu_1, \mu_2} \langle \mu_1 | \hat{\rho} | \mu_2 \rangle$$

where $\mu_1, \mu_2$ and $\mu_2$ are the Bohr magneton, the spin operator, and the radius vector of the $\mu$-th electron, while $\mu_2$ is the position of the $\mu$-th nucleus. The operator $T_1$ is given by

$$\frac{\partial}{\partial \mu_2} \langle \hat{\rho} \rangle = \sum_{\nu} \mu_2 \langle \nu | [\hat{\rho}, \hat{H}] | \mu_2 \rangle$$

where $\mu_2$ is the Hamiltonian of the electron subsystem. The symbol $\langle \rho \rangle$ (in $\mu_2$) denotes the trace over the electronic quantum numbers $\rho$, and $\nu$ is the density matrix of the electron subsystem.

Equation (1) is in fact contained in Abragam's book. It was derived by recognizing that the values of $\langle \hat{\rho} \rangle$ that play an important role in the influence of the form (1) are bounded by the correlation time of the orbital motion of the electron. The changes that occur during that time in the spin system can be neglected.

As already mentioned, the nuclear spin density matrix $\hat{\rho}$ can be assumed to be equivalent in equilibrium with a...
The average projection of the magnetic field direction in the stationary state is determined in terms of \( n_{\pm} = 1/\sqrt{2} \). Therefore:

\[
I_{\pm} = \frac{1}{\sqrt{2}} \left( n_{\pm} \frac{\partial}{\partial V_{\pm}} I_{\pm} \right) \tag{12}
\]

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 realize that our entire derivation is valid only if \( I_{\pm} \cong 1 \), since we use the high-temperature expansion of the nuclear spin density matrix. At \( \Delta \to 0 \), i.e., at thermal equilibrium, it follows from (11) that \( n_{\pm} = n \). The reason is that in the derivation of (4) we have in fact neglected the energy of the nuclear magnet in the magnetic field in comparison with \( T \). In fact, at \( \Delta \to 0 \) the value of \( n_{\pm} \) should, of course, tend to the lattice temperature \( T \).

The quantity \( I_{\pm} \) defined by (7) plays an important role in (11) only for a weak external field \( H \ll H_{0} \). In this case we have \( \mu_{\mp} \ll T \) and we can assume that \( H_{0} \) does not depend on the external field.

The form (11) yields the explicit dependence of \( n_{\pm} \) on the external field at a fixed value of \( \Delta \). As seen from this formula, the minimal value of the temperature reached is \( H \ll H_{0} \).

Here:

\[
\lim_{\Delta \to \infty} \frac{n_{\pm} - n_{\mp}}{n_{\pm} + n_{\mp}} = \frac{2}{\Delta} \tag{13}
\]

At \( \Delta \gg 1 \) it follows from (13) that \( n_{\pm} - n_{\mp} \ll 1/\Delta \) and strictly speaking formula (13) does not hold in this case, since the condition of the high-temperature approximation is violated. We can nevertheless expect \( \Delta \ll H \gg \Delta \gg 1 \).

4. The quantity \( I_{\pm} \) in (11) can be connected with the mean value of the electron spin projection \( S_{\mp} \) on the magnetic field direction. In particular, for nondegenerate electrons we have:

\[
\langle n_{\mp} \rangle = \frac{I_{\pm}}{2} = \frac{\langle S_{\mp} \rangle}{2}\tag{A.1}
\]

where \( S_{\mp} = -\frac{1}{2} (\Delta \mp (\Delta^{2} + 2)/2 \Delta \mp \Delta^{2}) \) is the equilibrium value of the electron spin, and \( \Delta \) is the electron g-factor in the semiconductors. Thus, for nondegenerate electrons formula (11) takes the form:

\[
I_{\pm} = \frac{2}{\sqrt{2}} \left( \frac{\partial}{\partial V_{\pm}} I_{\pm} \right) \tag{14}
\]

Of particular interest is the case when the electron spins are fully oriented \( \langle S_{\mp} \rangle = 0 \), i.e., in 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 \( I_{\pm} \ll \mu_{\mp} \), and formula (13) yields for the nuclear polarization:

\[
I_{\pm} = \frac{2}{\sqrt{2}} \left( \frac{\partial}{\partial V_{\pm}} I_{\pm} \right) \tag{15}
\]

This expression (so long as \( I_{\pm} \ll 1 \)) describes the Overhauser effect in arbitrary magnetic fields, including at \( H \ll H_{0} \).

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:

\[
I_{\pm} = \frac{2}{\sqrt{2}} \left( \frac{\partial}{\partial V_{\pm}} I_{\pm} \right) \tag{16}
\]

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