

THEORY OF NUCLEAR SPIN POLARIZATION INDUCED BY HOT ELECTRONS

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Nuclear spin polarization due to hot electrons (Feher effect) is studied theoretically for the case when the electron distribution function differs from a Boltzmann or Fermi distribution. The possible shape of the hot-electron distribution with respect to spin sublevels is investigated. The dependence of the nuclear polarization on the distribution function parameters is elucidated.

IN a "heated" system of conduction electrons, the distribution of the electrons with respect to their spin sublevels can greatly differ from the equilibrium distribution. If we assume for the time being that the electron distribution is described by a "kinetic" temperature T_e and that of the electron spins by a "spin" temperature T_s , then $T_e \neq T_s$ in the stationary state. In such a situation, all other spin subsystems of the crystal (nuclear spins, localized spins of the impurity atoms) will assume, by interaction with the conduction electrons, certain temperatures that differ from the lattice temperature. For the nuclear spins, owing to their very small relative specific heat, we can expect the strongest change in temperature. This phenomenon was predicted by Feher.^[1] Clark and Feher^[2] observed for the first time an increase in the polarization of nuclei interacting with the hot electrons.

From the theory of the Overhauser effect it is known that to produce nuclear polarization it is necessary that the polarization of the electron spins differ from the equilibrium polarization corresponding to the "kinetic temperature" T_e . In the Overhauser method this is attained by means of an external high frequency field, which depolarizes the electron spins, i.e., increases the "spin" temperature T_s . Owing to the large ratio of the magnetic moments of the electron and the nucleus, even even a small difference between the temperatures T_s and T_e produces a relatively large nuclear polarization. Exactly the same nuclear polarization, but with opposite sign, can be obtained by lowering the temperature of the electron spins relative to the "kinetic" temperature T_e . This is indeed realized in the Feher method, where the "kinetic" temperature is increased by direct current, and the "spin" temperature remains lower than the "kinetic" temperatures owing to the interaction between the spins and the lattice (for example,

with phonons or paramagnetic impurities).¹⁾

A theoretical examination of this phenomenon was made under the assumption that the non-equilibrium distribution of the conduction electrons is described by a Maxwellian or by a Fermi distribution function with effective temperatures T_e and T_s ^[1-6] and a drift velocity v .^[4-6] This assumption limits the region of applicability of the theory. We can present by way of an example a case (which is important in the sense of the magnitude of the effect), in which the formulas obtained earlier are not applicable directly. It is known that an increase in the polarization of the nuclei is determined by the difference in the temperatures $T_e - T_s$. This quantity depends strongly on the spin-lattice relaxation mechanism. Since the flipping of the electron spin is accompanied by a change in the electron energy by an amount $\hbar\omega_s$ (ω_s is the Zeeman frequency of the electron spin), we find that only a mechanism ensuring sufficiently high inelasticity of scattering can effectively separate the spin temperature T_s from the kinetic temperature T_e . This mechanism should at the same time make a sufficiently large contribution to the frequency of the spin-lattice relaxation. Estimates show^[6] that such a mechanism can be scattering by optical phonons. Simultaneously with spin relaxation, the optical phonons can determine also the electron energy relaxation and by the same token determine to a considerable degree the form

¹⁾In the investigation of the Overhauser effect in semiconductors, these two phenomena may become superimposed on each other. On the one hand, the external high-frequency field increases the spin temperature T_s directly, and on the other, the power absorbed from the field heats the electrons and, owing to the interaction between the spins and the lattice, the spin temperature is lowered relative to T_e . Since these phenomena "act in opposite directions," the resultant nuclear polarization may be smaller than expected.

of the distribution function, which frequently differs strongly in such cases from Maxwellian or a Fermi distribution.

We consider the dependence of the nonequilibrium nuclear polarization on the parameters of the distribution function, without making any assumptions concerning its explicit form.

1. We assume that the main contribution to the relaxation phenomena in the nuclei is made only by the contact part of the hyperfine interaction; then we can readily obtain, from the kinetic equations for the populations of the nuclear levels,^[7]

$$\begin{aligned} \langle I \rangle = & \frac{2I(I+1)}{3} \left\{ \sum_{\mathbf{k}\mathbf{k}'} |A|^2 [f(\mathbf{k}', -) - f(\mathbf{k}, +)] \right. \\ & \times \delta(\epsilon_{\mathbf{k},+} - \epsilon_{\mathbf{k}',-} - \hbar\omega_N) \left. \right\} \cdot \left\{ \sum_{\mathbf{k}\mathbf{k}'} |A|^2 [f(\mathbf{k}', -) \right. \\ & \times (1 - f(\mathbf{k}, +)) + f(\mathbf{k}, +) (1 - f(\mathbf{k}', -))] \\ & \left. \times \delta(\epsilon_{\mathbf{k},+} - \epsilon_{\mathbf{k}',-} - \hbar\omega_N) \right\}^{-1} \quad (1) \end{aligned}$$

where I is the spin of the nucleus and $\langle I \rangle$ is its mean value; $\hbar\omega_N$ is the Zeeman splitting of the energy levels of the nucleus; $\epsilon_{\mathbf{k},\sigma} = \epsilon_{\mathbf{k}} + \sigma\hbar\omega_S/2$ is the energy spectrum of the conduction electron; $|A|^2$ is the square of the matrix element of the contact interaction. This quantity does not depend on \mathbf{k} or \mathbf{k}' ; $f(\mathbf{k},\sigma)$ is the distribution function of the conduction electrons.

Assuming first that the spectrum $\epsilon(\mathbf{k})$ is isotropic, we represent the nonequilibrium function in the form of a series of spherical functions:

$$f(\mathbf{k},\sigma) = \sum_{nl} [F_{nl}(\epsilon) + \sigma\Phi_{nl}(\epsilon)] Y_{nl}(\theta, \varphi). \quad (2)$$

We disregard here phenomena connected with the change in the electron spectrum in external fields. We are likewise not interested in the off-diagonal elements of the density matrix $f_{\sigma\sigma'}$, since, to the extent that relations (1) are valid, the polarization of the nuclear spins is determined only by the diagonal elements.

Since the square of the matrix element $|A|^2$ does not depend on the angles ϑ or φ and the spectrum is isotropic, we can readily see that $\langle I \rangle$ is determined only by two terms of the series (2): $F_0(\epsilon)$ and $\Phi_0(\epsilon)$. Integrating over the angles in (1) and confining ourselves to terms of lowest order in $\hbar\omega_S/\bar{\epsilon}$ ($\bar{\epsilon}$ is the average energy of the conduction electrons), we obtain

$$\langle I \rangle \cong \frac{I(I+1)}{3} \left\{ \int_0^\infty d\epsilon d\epsilon' \rho(\epsilon) \rho(\epsilon') \delta(\epsilon - \epsilon' + \hbar(\omega_S - \omega_N)) \right.$$

$$\begin{aligned} & \left. \times [F_0(\epsilon') - F_0(\epsilon) - \Phi_0(\epsilon') - \Phi_0(\epsilon)] \right\} \\ & \times \left\{ \int_0^\infty d\epsilon d\epsilon' \rho(\epsilon) \rho(\epsilon') F_0(\epsilon) [1 - F_0(\epsilon')] \delta(\epsilon - \epsilon') \right\}^{-1}. \end{aligned}$$

Here $\rho(\epsilon)$ is the state density of the conduction electrons.

We shall henceforth find it more convenient to consider the ratio $\langle I \rangle / \langle I \rangle_0$, where $\langle I \rangle_0$ is the average spin of the nucleus in the absence of external fields at a lattice temperature T_0 . In this case

$$\begin{aligned} \frac{\langle I \rangle}{\langle I \rangle_0} = & - \left\{ \int_0^\infty d\epsilon \rho^2(\epsilon) \frac{\partial F_0}{\partial \epsilon} T_0 \right\} \\ & \times \left\{ \int_0^\infty d\epsilon \rho^2(\epsilon) F_0(\epsilon) [1 - F_0(\epsilon)] \right\}^{-1} + \frac{\gamma_e}{\gamma_N} \left\{ \int_0^\infty d\epsilon \rho^2(\epsilon) \right. \\ & \times T_0 \left[\frac{\partial F_0}{\partial \epsilon} - (\hbar\omega_S)^{-1} \Phi_0(\epsilon) \right] \left. \right\} \\ & \times \left\{ \int_0^\infty d\epsilon \rho^2(\epsilon) F_0(\epsilon) [1 - F_0(\epsilon)] \right\}^{-1} \quad (3) \end{aligned}$$

(γ_e and γ_N are the gyromagnetic ratios of the electrons and of the nucleus, respectively). Formula (3) is valid also for sufficiently fast high-frequency processes, when the functions $F_0(\epsilon)$ and $\Phi_0(\epsilon)$ do not depend on the time.

The functions $F_0(\epsilon)$ and $\Phi_0(\epsilon)$ are determined from the kinetic equation with account of collisions with spin flip. Since the probabilities of the transition with spin flip are usually much lower than the corresponding probabilities of transition without spin flip, the function $\Phi_0(\epsilon)$ is determined in the zeroth approximation in their ratio in terms of $F_0(\epsilon)$, accurate to an indeterminate constant. Indeed, the kinetic equations for the determination of the function $f(\mathbf{k},\sigma)$ have in this approximation the form

$$\hat{L}f_\sigma(\mathbf{k}) = J(f_\sigma(\mathbf{k})). \quad (4)$$

Here \hat{L} is the Liouville operator, $J(f_\sigma(\mathbf{k}))$ is the collision integral, in which only the relaxation mechanisms without spin flip are taken into account.

The integro-differential equations (4) determine the functions $f_\sigma(\mathbf{k})$ accurate to two indeterminate constants C_+ and C_- . In the lowest approximation in $\hbar\omega_S/\bar{\epsilon}$, the functions $F_0(\epsilon)$ and $\Phi_0(\epsilon)$ then take the form

$$\begin{aligned} F_0(\epsilon) = & {}^{1/2}[f_0(\epsilon, C_+) + f_0(\epsilon, C_-)] \cong f(\epsilon, z), \quad (5) \\ \Phi_0(\epsilon) = & \frac{1}{2}[f_0(\epsilon, C_-) - f_0(\epsilon, C_+)] \end{aligned}$$

$$\cong \frac{C_- - C_+}{2} \frac{\partial f_0(\varepsilon, z)}{\partial z} \equiv \frac{\hbar\omega_s}{2T_s} \frac{\partial F_0}{\partial z}. \quad (6)$$

Here $f_0(\varepsilon, C_{\pm})$ are the symmetrical parts of the function $f_{\pm}(\mathbf{k})$; we put for convenience

$$\frac{C_+ + C_-}{2} = z, \quad \frac{C_- - C_+}{2} = \frac{\hbar\omega_s}{2T_s}.$$

It is easy to see that actually $C_- - C_+ \sim \hbar\omega_s/T_s \ll 1$. The new indeterminate constant z is determined, for example, from the normalization condition (or from the conservation of the particle-number density, if the particle number is not constant). The constant T_s (which has the dimension of energy), is determined from the condition for the conservation of the spin density in collisions and depends already on the mechanism of spin-lattice relaxation. For a Fermi or Maxwellian function $F_0(\varepsilon)$, the quantity T_s has the meaning of a spin temperature. Thus, the increase in the nuclear polarization $\langle I \rangle / \langle I \rangle_0$ is determined only by a single function $F_0(\varepsilon, z)$.

If we neglect the Pauli principle and electron-electron collisions in the collision integral (4), then the equations become linear and determine $F_0(\varepsilon, z)$ accurate to an arbitrary factor.²⁾ In this case

$$\Phi_0(\varepsilon) = \frac{\hbar\omega_s}{2T_s} F_0(\varepsilon), \quad (7)$$

$$\frac{\langle I \rangle}{\langle I \rangle_0} = \frac{T_0}{M} \left[1 + \frac{\gamma_e}{\gamma_N} \left(\frac{M}{T_s} - 1 \right) \right], \quad (8)$$

$$\begin{aligned} \frac{1}{M} &= - \int_0^{\infty} d\varepsilon \rho^2(\varepsilon) \frac{\partial F_0}{\partial \varepsilon} \bigg/ \int_0^{\infty} d\varepsilon \rho^2(\varepsilon) F_0(\varepsilon) \\ &= \int_0^{\infty} d\varepsilon \left(\frac{\partial}{\partial \varepsilon} \rho^2(\varepsilon) \right) F_0(\varepsilon) \bigg/ \int_0^{\infty} d\varepsilon \rho^2(\varepsilon) F_0(\varepsilon). \end{aligned} \quad (9)$$

For a parabolic spectrum we have

$$M = \int_0^{\infty} d\varepsilon \varepsilon F_0(\varepsilon) \bigg/ \int_0^{\infty} d\varepsilon F_0(\varepsilon). \quad (10)$$

It is easy to see that if $F_0(\varepsilon)$ is a Maxwellian function with effective temperature T_e , then $M = T_e$ and formula (8) goes over into the formula previously derived by Feher.^[1]

When account is taken of the Pauli principle under sufficiently general assumptions concerning the electron relaxation mechanisms, the symmetrical part of the distribution function can be represented in the form^[8]

$$F_0(\varepsilon) = [1 + e^{z(\varepsilon)-z}]^{-1}, \quad (11)$$

where $z(\varepsilon)$ is a function of the energy, whose explicit form is determined by the relaxation mechanisms. According to (6)

$$\Phi_0(\varepsilon) = \frac{\hbar\omega_s}{2T_s} F_0(\varepsilon) [1 - F_0(\varepsilon)], \quad (12)$$

and the formula for the increase in the nuclear polarization will also have the same form as (8), where M must be set equal to

$$M = \int_0^{\infty} d\varepsilon \rho^2(\varepsilon) \frac{\partial F_0}{\partial z} \bigg/ \int_0^{\infty} d\varepsilon \rho^2(\varepsilon) \frac{\partial F_0}{\partial \varepsilon}. \quad (13)$$

Naturally, $M = T_e$ if $F_0(\varepsilon)$ is a Fermi distribution with effective temperature T_e . For the case of strong degeneracy, it is easy to obtain a simpler formula for M . In this case the function $\partial F_0 / \partial z$ can be approximately replaced by $\delta(z(\varepsilon) - z)$; then

$$M = (d\varepsilon / dz)_{z(\varepsilon)=z}. \quad (14)$$

2. We present also formulas for $\langle I \rangle / \langle I \rangle_0$ for crystals with a complicated energy spectrum. We assume that the electrons are distributed near the extrema of the energy band. For integration in (1), we use the well known formula

$$\sum_{\mathbf{k}} \psi_{\mathbf{k}} = \frac{1}{(2\pi\hbar)^3} \sum_{\alpha} \int d\varepsilon_{\alpha} \int \frac{dS^{\alpha}}{|\nabla_{\mathbf{p}} \varepsilon_{\alpha}|} \psi_{\mathbf{k}}. \quad (15)$$

Here ϑ^{α} and φ^{α} are the polar and azimuthal angles of the vector $\mathbf{k} - \mathbf{k}_{\alpha}$; \mathbf{k}_{α} is the extremal point of the energy band (α is the number of the valley).

We shall consider the contribution made only by the zeroth terms of the series (2). Under the same assumptions as used in the derivation of (11) and (12), we can show that

$$F_0^{\alpha}(\varepsilon) = [1 + e^{z(\varepsilon)-z_{\alpha}}]^{-1}, \quad \Phi_0^{\alpha}(\varepsilon) = \frac{\hbar\omega_s^{\alpha}}{2T_s^{\alpha}} \frac{\partial F_0^{\alpha}}{\partial z_{\alpha}}. \quad (16)$$

Here z^{α} and T_s^{α} are obtained from the system of equations for the conservation of the particle-number density and the spin density with account of the intervalley transitions.

Neglecting the intervalley transitions in the scattering of the electron by the nuclear spin and carrying out the integration in (1), we obtain

$$\frac{\langle I \rangle}{\langle I \rangle_0} = \frac{A_1}{A_0} + \frac{\gamma_e}{\gamma_N} \left(\frac{A_2}{A_0} - \frac{A_3}{A_0} \right);$$

$$A_0 = \sum_{\alpha} |A|_{\alpha}^2 \int_0^{\infty} d\varepsilon (\rho_0^{\alpha}(\varepsilon))^2 \frac{\partial F_0^{\alpha}}{\partial z_{\alpha}},$$

$$A_1 = \sum_{\alpha} |A|_{\alpha}^2 \int_0^{\infty} d\varepsilon (\rho_0^{\alpha}(\varepsilon))^2 \frac{\partial F_0^{\alpha}}{\partial \varepsilon} T_0,$$

²⁾This conclusion remains valid also in the limiting case of strong electron-electron interaction.

$$A_2 = \sum_{\alpha} \frac{\gamma_{\alpha}}{\gamma_e} |A|_{\alpha}^2 \int_0^{\infty} d\varepsilon (\rho_0^{\alpha}(\varepsilon))^2 \frac{\partial F_0^{\alpha}}{\partial \varepsilon} T_0,$$

$$A_3 = \sum_{\alpha} \frac{\gamma_{\alpha}}{\gamma_e} |A|_{\alpha}^2 \int_0^{\infty} d\varepsilon (\rho_0^{\alpha}(\varepsilon))^2 \frac{\partial F_0^{\alpha}}{\partial z_{\alpha}}. \quad (17)$$

Here $|A|_{\alpha}^2$ is the square of the matrix element of the contact interaction for the α -valley. The difference in the gyromagnetic ratios γ_{α} for different valleys can be attributed, for example, to the anisotropy of the conduction-electron g-tensor. Further,

$$\rho_0^{\alpha}(\varepsilon) = \int \frac{dS^{\alpha}}{|\nabla_{\mathbf{p}} \varepsilon_{\alpha}|}.$$

3. We make a few concluding remarks. Owing to the large value of the ratio γ_e/γ_N ($\sim 10^3-10^5$), the increase in the polarization $\langle I \rangle / \langle I \rangle_0$ can serve as a very sensitive method for measuring the difference between M and T_S , which is of interest for the study of spin-lattice relaxation of nonequilibrium carriers in crystals. If we measure T_S independently, for example using the spin-resonance signal, then the ratio $\langle I \rangle / \langle I \rangle_0$ determines the quantity M , which characterizes the distribution $F_0(\varepsilon)$. In conjunction with methods which give other moments of the distribution function, the increase in the nuclear polarization can be used to reconstruct the form of the distribution function. When making concrete use of formulas (3), (8), and (17), it is necessary to bear in mind also other mechanisms of relaxation of nuclear spins, which lead to "leakage" of the polarization.^[7]

It follows from the derivation of formulas (3) in (8) that the carrier drift and other singularities of the carrier motion, described by the higher terms of the series (2), do not make any contribution to the nuclear polarization in the case of an isotropic spectrum. The contribution of the drift to $\langle I \rangle$, which was considered in^[3, 5, 6], is a result of an inaccurate determination of the effective temperatures T_e and T_S .³⁾ We note that in anisotropic crystals without an inversion center, the con-

tribution to the nuclear polarization can be made also by the asymmetrical part of the distribution function, which we discarded in the derivation of formula (17). It is possible that this can explain the contribution observed in^[2] to the nonequilibrium polarization, which is proportional to the components of the vector $\mathbf{H} \times \mathbf{J}$, where \mathbf{H} and \mathbf{J} are the constant magnetic field and the electric current in the sample.

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¹G. Feher, Phys. Rev. Lett. **3**, 135 (1959).

²W. G. Clark and G. Feher, Phys. Rev. Lett. **10**, 134 (1963).

³M. Weger, Phys. Rev. **132**, 581 (1963).

⁴L. L. Buishvili, FTT **6**, 108 (1964), Soviet Phys. Solid State **6**, 85 (1964).

⁵V. P. Kalashnikov, FTT **6**, 2457 (1964) and **7**, 3180 (1965), Soviet Phys. Solid State **6**, 1949 (1965) and **7**, 2540 (1966).

⁶A. K. Zvezdin, JETP **49**, 1313 (1965), Soviet Phys. JETP **22**, 907 (1966).

⁷A. Abragam, The Principles of Nuclear Magnetism, Oxford, 1961.

⁸V. P. Shabanskiĭ, JETP **27**, 142 and 150 (1954).

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³⁾In^[3,5,6] the distribution function was taken in the form

$$f \left[\frac{\varepsilon(\mathbf{p} - m\mathbf{V}) + \sigma \hbar \omega_s / 2 - \zeta_{\sigma}}{T_e} \right].$$

If we expand it in the series (2) and redefine T_e and T_S , as usual, in terms of the zero-order function $F_0(\varepsilon)$ and $\Phi_0(\varepsilon)$, then the formulas from^[3,5,6] will coincide with (8) when $M = T_{\text{eff}}$, and the carrier drift proper will make no contribution to the nuclear polarization.