Phase transition in a nuclear spin system in a semiconductor with optically oriented electrons

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The spin temperatures and polarization of nuclei in a semiconductor with oriented electrons are calculated theoretically by assuming that the spin–spin interaction between neighboring nuclei is mainly scalar, and that the correlation radius of the electron orbital motion is large compared with the lattice constant. It is shown that in this case the spin temperature can be found without resorting to the high-temperature approximation. An analysis of the results shows that by means of optical orientation the spin-system temperature can be made lower than the temperature of the phase transition due to the scalar interaction between the nuclear spins.

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1. When the electrons of a semiconductor are optically oriented in a magnetic field, the spin system of the lattice nuclei is cooled.[1,2] D'yakonov and Perel'[1] obtained a kinetic equation for the nuclear density matrix and found the nuclear spin temperature (θ) for the case when it is high compared with the characteristic energy of the nuclear spin. If the high-temperature approximation is not valid, the determination of θ entails as a rule considerable difficulties and has not been performed to data.

Yet this spin-temperature range is of particular interest. If θ is of the order of the energy of the spin-spin interaction between the lattice nuclei, a phase transition should take place in the nuclear spin system. The spins of the lattice nuclei become ordered.[3] By solving the problem outside the region where the high-temperature approximation is valid it would be possible to determine whether a phase transition can be produced in the spin system of the semiconductor nuclei when the electrons are oriented. (It follows from the high-temperature calculations[1] that if a magnetodipole interaction takes place between the spins of neighboring nuclei, the necessary condition for cooling below the phase-transition temperature is a high polarization of the oriented electrons.)

We show in the present paper that if the spin-spin interactions between neighboring nuclei are mainly scalar, the temperature of the nuclear spin system can be obtained without using the high-temperature approximation.[1)] From an analysis of the results it follows that in this case the spin system of the lattice nuclei can be cooled to below the phase-transition temperature. The cooling must be carried out in a weak magnetic field H, weaker than the local field H_L produced at the nucleus by the neighboring nuclei on account of the scalar interaction. The smaller the polarization of the oriented electrons, the weaker must H be.

2. We assume that the spin-spin interactions between neighboring nuclei are mainly scalar[5]:

$$\hat{H}_{II}=\sum_{mn} A_{mn}(\hat{\mathbf{I}}_m\hat{\mathbf{I}}_n), \qquad (1)$$

where $\hat{\mathbf{I}}_m$ and $\hat{\mathbf{I}}_n$ are the spin operators of the nuclei numbered m and n respectively, while the coefficients A_{nm} depends only on the positions of these nuclei. The Hamiltonian (1) commutes with the projection operators of the total spin of the lattice nuclei. Thus, in a constant magnetic field H directed along the Z axis the total angular momentum (I) and its projection on the Z axis (M) are "good" quantum numbers.

We shall regard the dipole-dipole interaction between neighboring nuclei as a small perturbation that leads to establishment of a thermodynamic-equilibrium distribution over the nuclei spin levels with different values of I, M, and other quantum numbers which we shall designate for brevity by a single letter γ. We neglect the influence of the dipole-dipole interaction on the form of the wave functions of the levels of the nuclear spin systems. This is valid if the external magnetic field exceeds the characteristic field H_{LD} connected with the dipole interaction between neighboring nuclei ($H_{LD} \ll H$), and H_{LD} is much smaller than the local field H_L due to the scalar interaction.

We examine now how such a nuclear-spin system is cooled by oriented electrons.

A distinguishing feature of the polarization of nuclei by electrons in a semiconductor is that the electron interacts simultaneously with a large number of nuclear spins. (The radius of the correlations of the orbital motion of the electrons if large compared with the lattice constant.) We assume that all the lattice nuclei are identical, so that the hyperfine contact interaction does not depend on the number of the nucleus. It can then be assumed with sufficient accuracy (see the Appendix) that at each instant of time the electron interacts with the total nuclear spin **I**:

$$\hat{V}=a\sum_i(\hat{\mathbf{S}}_i,\hat{\mathbf{I}})=a\sum_i\left(\hat{\mathbf{S}}_i,\ \sum_n\hat{\mathbf{I}}_n\right), \qquad (2)$$

where $\hat{\mathbf{S}}_i$ is the spin operator of the i-th electron. This interaction produces transitions only between states with identical quantum numbers I and γ. (The projection M changes in one transition by unity, $M \to M \pm 1$.) Thus, the problem of the polarization of all the nuclei turns out to be equivalent to the problem of dynamic polarization of one nucleus with angular momentum I

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(Ref. 6).

We recall now the reasoning used to solve this problem. Let the population of the nuclear spin levels with quantum numbers M, I, and γ be described by a certain spin temperature θ. We then have for the occupation numbers

$$\frac{N(M+1,I,\gamma)}{N(M,I,\gamma)}=\exp\left(\frac{\mu_j}{j}\frac{M}{\theta}\right), \qquad (3)$$

where μ_j and j are the magnetic moment and the spin of the lattice nucleus.

On the other hand, since the systems of nuclear and electron spins are in dynamic equilibrium, we have

$$N(M+1,\ I,\ \gamma)n(-{}^1/_2)W_{\uparrow\downarrow}=N(M,\ I,\ \gamma)n({}^1/_2)W_{\downarrow\uparrow}. \qquad (4)$$

Here $n(1/2)$ and $n(-1/2)$ are the numbers of electrons with spin parallel and antiparallel to the magnetic field H, while $W_{\uparrow\downarrow}$ and $W_{\downarrow\uparrow}$ are the respective probabilities of the transitions $(-1/2;M+1,I,\gamma)\rightarrow(1/2;M,I,\gamma)$ and $(1/2;M,I,\gamma)\rightarrow(-1/2;M+1,I,\gamma)$. From the detailed-balancing principle we have

$$W_{\uparrow\downarrow}/W_{\downarrow\uparrow}=\exp\{(-\mu_jH/j+\mu gH)/T\}, \qquad (5)$$

where μ and g are the Bohr magneton and the g-factor of the electron, and T is the lattice temperature. Therefore

$$\frac{N(M+1,I,\gamma)}{N(M,I,\gamma)}=\frac{n({}^1/_2)}{n(-{}^1/_2)}\exp\left\{\left(\frac{\mu_j}{j}H-\mu gH\right)\Big/T\right\}. \qquad (6)$$

If the electrons are polarized, then

$$\frac{n({}^1/_2)}{n(-{}^1/_2)}=\exp\left\{\frac{\mu gH+\Delta\zeta}{T}\right\}. \qquad (7)$$

Here $\Delta\zeta=\zeta_+-\zeta_-$ is the difference between the chemical potentials of the electrons with the spins parallel and antiparallel to the magnetic field, and is connected with the average electron spin $(\bar{S}z)$ and its equilibrium component in the field $H(S_T)$ by the relation[1]

$$\mathrm{th}\frac{\Delta\zeta}{2T}=\frac{2(\bar{S}_z-S_T)}{1-4S_zS_T}\approx2\bar{S}_z. \qquad (8)$$

Substituting (3) and (7) in (6) we obtain the final expression for the spin temperature of the nuclei:

$$\frac{1}{\theta}=\frac{1}{T}+\frac{j}{\mu_j}\frac{\Delta\zeta}{HT}. \qquad (9)$$

As is easily seen, it follows from this expression that the nuclear spin system can be cooled below the phase-transition temperature (θ_c). Indeed, if the electron polarization is high enough ($\Delta\zeta\sim T$), when $\theta\sim(\mu_j/j)H$. The nuclear spins become ordered at a temperature θ_c of the order of the spin-spin scalar interaction parameter A, so that at magnetic field values

$$H<\frac{j}{\mu_j}\frac{\Delta\zeta\theta_c}{T}\approx\frac{j}{\mu_j}\frac{\Delta\zeta A}{T} \qquad (10)$$

the spin-system nuclear temperature turns out to be lower than θ_c.[2)]

As shown above, this conclusion follows unambiguously from the assumption that the spin system of the nuclei (described by the temperature θ) is in a quasi-equilibrium state and from the form of the stationary distribution of the total nuclear spin over the sublevels with different values of M. The surprising fact is that the distribution both above and below the phase-transition temperature is given by one and the same equation (3) which contains no singularities whatever connected with the ordering of the nuclear spins. It might seem at first glance that the nuclear polarization should likewise not undergo substantial changes when the temperature is decreased below θ_c. This, however, is naturally not the case. The point is that at temperatures lower than θ_c a substantial change takes place in the distribution over states with different values of I and γ.

In the ferromagnetic phase, for example, the most probable are the states with $I\sim Nj$, where N is the number of nuclei in the lattice and j is the angular momentum of one nucleus. At one and the same ratio $N(M+1)/N(M)$, larger values of I lead to considerably larger values of the polarization. In a transition into the ferromagnetic state, therefore, the polarization of the nuclear spins should be substantially larger than their polarization in the high-temperature region (see below).

3. We shall calculate the nuclear polarization in the simplest model of the self-consistent Weiss field.[7] We shall assume that the nuclear spin is acted upon, besides the external field H, by the Weiss field $H_W=\lambda\langle I\rangle$, which is connected with the scalar interaction between the neighboring nuclei and is proportional to the average spin $\langle I\rangle$ of the lattice nuclei. Here λ is a proportionality coefficient equal in order of magnitude to Aj/μ_j. The average nuclear spin is then the solution of the equation

$$\langle I\rangle=\left(j+\frac{1}{2}\right)\mathrm{cth}\left[\frac{\mu_j(2j+1)}{2j\theta}(H+\lambda\langle I\rangle)\right]-\frac{1}{2}\mathrm{cth}\left[\frac{\mu_j}{2j\theta}(H+\lambda\langle I\rangle)\right]. \qquad (11)$$

Using the expression (9) for the nuclear spin temperature, in which we neglect for simplicity the term $1/T$, we easily obtain the final equation for the polarization of semiconductor nuclei upon optical orientation of the electrons. In particular, if the nuclear spin is 1/2

$$\langle I\rangle=\frac{1}{2}\mathrm{th}\left[\frac{\Delta\zeta}{2T}\left(1+\frac{\lambda\langle I\rangle}{H}\right)\right]. \qquad (12)$$

At sufficiently low values of the external magnetic field, when the Weiss field is comparable with or exceeds H, this equation for $\langle I\rangle$ can be solved only numerically. It is easy, however, to solve with the aid of (12) the inverse problem, namely, given the values of $\langle I\rangle$ and $\Delta\zeta/T$, determine the field H that satisfies this equation, namely

$$H=\lambda\langle I\rangle\left[\frac{2T}{\Delta\zeta}\ln\left(\frac{1+2\langle I\rangle}{1-2\langle I\rangle}\right)-1\right]^{-1}. \qquad (13)$$

The results of the calculations by means of this formula, at a given average spin $\bar{S}_z\approx\Delta\zeta/4T=0.5$ of the oriented electrons, are shown in Fig. 1. (The magnetic field is measured in units of the Weiss-field parameter λ.)

It is seen that at large value of the magnetic field the polarization of the lattice nuclei is practically independent of H and does not change when the sign of the field is reversed. This is the usual consequence of the high-temperature approximation, where $\mu_j H/\theta \sim \langle I \rangle \approx \bar{S}_z$.

In weak fields, a phase transition takes place in the nuclear-spin system. At positive values of the ratio H/λ the system goes over into the ferromagnetic state and the degree of polarization ($\langle I \rangle/j$) tends to unity. On the other hand, if $H/\lambda < 0$, the nuclear spins are antiferromagnetically ordered and the mean value of the nuclear spin tends to zero as $H \to 0$.

4. It follows from the foregoing analysis that if the interaction between the spins of the neighboring nuclei of the semiconductor is mainly scalar, electron orientation in a weak magnetic field can lead to a phase transition in a nuclear spin system. The spin system is cooled into the region of positive or negative temperatures, depending on whether the electrons are polarized parallel or antiparallel to the magnetic field. The ordering of the nuclear spins is then ferromagnetic or antiferromagnetic.

In conclusion, we emphasize once more that in the model considered the thermodynamic-equilibrium distribution over the nuclear-spin level is established because of the dipole-dipole interaction between the spins of neighboring lattice nuclei. Without this interaction, the state of the nuclear system is not described by a single parameter (temperature) and no phase transition can occur, since the principal scalar interaction conserves the total nuclear spin and cannot lead to a predominant population of states with larger values of the total spin.

At the same time, an important factor in the entire analysis is the smallness of the dipole-dipole interaction. This analysis is therefore valid only if the characteristic relaxation time of the nuclear spins on the electrons is longer than the long time T_2 during which the dipole interaction establishes a thermo-dynamic-equilibrium distribution over the nuclear spin levels.

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APPENDIX

In the derivation of the expression for the spin temperature we have assumed that the correlation radius (r_c) of the orbital motion of the electrons is infinitely large, i.e., it was assumed that the electron interacts with the total spin of the lattice nuclei. If this radius is finite, the electron interacts simultaneously only with nuclei separated by distances $r < r_c$. The total spin of such nuclei, is acted upon, besides by the external field H, also by the random field H'_L due to scalar interaction with other spins. This local field must also be taken into account when the spin temperature is calculated. However, since the spin-spin interactions are significant only between nuclei that are separated by a distance not exceeding several lattice constants (b), while the correlation radius in the semicondutor is large compared with b, the field H'_L can be neglected if

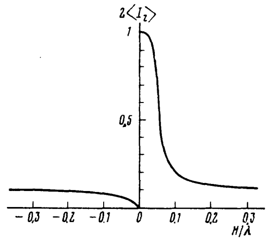

FIG. 1. Dependence of the degree of polarization of the lattice nuclei of a semiconductor on the magnetic field in which the cooling takes place. The rapid increase of the polarization at small positive values of the magnetic field is due to the phase transition into a ferromagnetic state, and the decrease of the polarization at small negative values of H/λ is due to the antiferromagnetic ordering of the nuclear spins.

$$4\pi r_c^2 b|A| \ll 4\pi r_c^3 \mu_j H/(3j).$$

In the derivation of (9) we have also disregarded the change of the nuclear-system wave functions on account of the dipole-dipole interaction. This approximation is justified if the energy of the dipole interaction between the neighboring nuclei ($E_d \sim \mu_j^2/b^3$) is small compared with the energy of their spins in the magnetic field and with the energy of the scalar spin-spin interaction. The expression obtained for the spin temperature of the semiconductor-lattice nuclei is therefore valid in a region where the lower bound of the magnetic field is given by the inequality

$$\mu_j H \gg \max\{|A| b/r_c;\ \mu_j^2/b^3\}.$$

This condition can also be obtained by solving the kinetic equation for the spin density matrix of the lattice nuclei.[1]

1) This interaction is due to the indirect exchange via the electrons and it is sometimes called exchange interaction.[4]

2) We recall that in the derivation of (9) we have assumed that the correlation radius of the orbital motion of the electrons is infinite, and that the dipole-dipole interaction between spins of neighboring nuclei is negligibly small. As shown in the Appendix, these assymptions impose a lower bound on the field H.

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