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The limiting nuclear polarization in a quantum dot under optical electron-spin orientation and applicability of the box-model of the electron-nuclear dynamics.

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For the model Hamiltonian describing the electron-nuclear dynamics of a quantum dot, we obtained an exact expression for the limiting nuclear polarization as a function of the number of groups of equivalent nuclei. It is shown that the refinement of the model Hamiltonian by increasing the number of the groups results in a slow growth of the limiting nuclear polarization. This allowed us to put forward arguments in favor of applicability of the box-model (with all the nuclei being equivalent) for description of the electron-nuclear spin dynamics within the time intervals of around hundreds of periods of the optical orientation.

I. INTRODUCTION

Spin systems involving electron spin coupled by contact interaction with a large number of nuclear spins are well known in radio-spectroscopy (rf spectroscopy). The appropriate quantum-mechanical models are well developed for the case of large magnetic fields typical for the rf spectroscopy (when the Zeeman energy of an electron spin in an external magnetic field substantially exceeds the energy of electron-nuclear interaction). In recent years, there have appeared new objects – quantum dots, whose electron-nuclear dynamics can be described by similar models [1].

A typical experiment on spin dynamics of a quantum dot implies observation of time dependence of spin of an electron localized in a quantum dot initially spin-oriented by a short polarized laser pulse. Observation of the electron spin dynamics, in these experiments, is performed within the time intervals when the contact interaction of this spin with nuclear spins of the quantum dot appears to be predominant. The experiments are usually interpreted using the Hamiltonian

$$H = \omega S_z + \sum_i \left\{ A_{i\parallel} S_z I_{zi} + A_{i\perp} [S^+ I_i^- + S^- I_i^+] \right\}$$

(1)

where $\omega$ is the external magnetic field in frequency units, and $A_{i\parallel}$ and $A_{i\perp}$ – are tensor components of the hyperfine interaction between the i-th nuclear and the electron spin, $S_z$ ($I_{zi}$) – are the operators of the z-projection of the electron spin (i-th nuclear spin), and $S^z$ ($I_i^\pm$) are the electron (nuclear) spin z-projection rising and lowering operators. In contrast to the radio-spectroscopic experiments, these experiments are carried out in a wide range of magnetic fields. For this reason, theoretical methods of rf spectroscopy cannot be used to analyze experiments with quantum dots. A great number of nuclei interacting with the electron spin makes it impossible to solve appropriate problems numerically using computers. This is why exactly solvable models of the electron-nuclear spin dynamics are of particular interest even when the exact solution is obtained at the expense of assumptions whose plausibility cannot be reliably evaluated.
In the theory of spin dynamics of the quantum dots, an exact solution can be obtained in the framework of the box-model \[2, 3, 4\], which implies that the electron spin is coupled in the same way with all the nuclear spins of the quantum dot. Thus, the box-model postulates that the electron density (electron wavefunction module squared) is constant within the quantum dot. The calculations of the electron-nuclear spin dynamics fulfilled in the framework of the box-model have shown its high efficiency. With the aid of the box-model, it became possible: (i) to describe electron spin dynamics after optical orientation, (ii) to obtain the magnetic-field dependence of the residual electron polarization of the quantum dot, (iii) to show that, in the experiments on spin dynamics of quantum dots, one should expect a strong deviation of the nuclear state from equilibrium, (iv) to describe the effect of replicas arising in the electron spin dynamics (the effect results from appearance of regularity in the nuclear density matrix under condition of periodic optical orientation of the electron spin), and (v) to predict the echo-effect in spin dynamics of a single quantum dot after a \(\pi\)-pulse of the magnetic field. These facts show that the simplifications laid down into the basis of the box-model are not important for description of a number of experiments.

The box-model, however, has at least one property that restricts its application and needs to be additionally discussed. The point is that, in the framework of the box-model, it is impossible to obtain a high nuclear polarization for the constant-sign orientation of the electron spin. For quantum dots with \(10^4\) nuclei, the limiting polarization calculated in the framework of the box-model lies in the range of 1\%, whereas there have been reported experimental observations of the nuclear polarization of several tens of percents \[5, 6, 7\]. The above properties of the box-model are related to the fact that all nuclei of the quantum dot, in this model, are considered to be equivalent (with respect to interaction with the electron spin localized in the quantum dot) and the Hamiltonian of the electron-nuclear interaction depends on the total nuclear moment. In real systems, the electron density is not constant over the quantum dot. This fact violates the restriction imposed on the limiting nuclear polarization following from the box-model.

The goal of the present work is to study dependence of the limiting nuclear polarization arising in the quantum dot upon optical orientation of the electron spin on the degree of nonuniformity of the electron density in the quantum dot. The spatial nonuniformity of electron density in the quantum dot can be taken into account by characterizing this nonuniformity by surfaces of equal density and assuming that the nuclei located between the neighboring surfaces are equivalent with respect to their coupling to the electron spin in the quantum dot. To be more precise, we will choose, in the range of variation of the electron density, \(n\) levels \(|\Psi_i|^2, i = 1, \ldots, n\), so that: \(0 = |\Psi_0|^2 < |\Psi_1|^2 < |\Psi_2|^2 < \ldots < |\Psi_n|^2 \equiv |\Psi|_{\text{max}}^2\). The nuclei of the region with electron density \(|\Psi|^2\) lying in the range \(|\Psi_{i-1}|^2 < |\Psi|^2 < |\Psi_i|^2\) are referred to \(i\)-th group and are considered to be equally coupled to the electron. In this way, we obtain \(n\) groups of equivalent nuclei. The accuracy of such a description will improve with increasing number of the groups. The box-model evidently corresponds to the case of \(n = 1\). The main result of this paper is the exact expression for the limiting nuclear polarization of the electron-nuclear system with a given number \(n\) of the groups of equivalent nuclei and with a given number of nuclei in each group. For large number of nuclei in the groups, the limiting polarization appears to be higher than that in the box-model by a factor of \(\sqrt{n}\) as the upper limit.
II. THE LIMITING NUCLEAR POLARIZATION FOR A GIVEN NUMBER OF GROUPS OF EQUIVALENT NUCLEI

In what follows, we will consider the electron-nuclear spin system with the elementary nuclear angular momenta equal to $\frac{1}{2}$. Recall that in this case the expression for the limiting nuclear polarization $P_1$ for the box-model with the number of equivalent nuclei equal to $2N$, has the form [4]:

$$P_1 = P(N) = \frac{1}{2N} \frac{1}{2^{2N}} \sum_{J=0}^{N} \Gamma_N(J)(2J+1)J \approx \frac{1}{\sqrt{N\pi}}$$

where

$$\Gamma_N(J) = C_{2N}^{N-J} - C_{2N}^{N-J-1}, \quad J = 0, 1, \ldots, N$$

The approximate equality is valid for large $N$.

Now, we will extend the box-model in the following way. We will assume that the spin system has two, rather than one, group of the equivalent nuclei characterized by two tensor constants of contact interaction $A_1$ and $A_2$ and containing, respectively, $2N_1$ and $2N_2$ nuclear spins. The relevant Hamiltonian written in frequency units will be given by:

$$H_2 = \omega S_z + H_1 + H_2$$

where the first term corresponds to Zeeman energy of the electron spin in the external magnetic field $\omega$, and the operators

$$H_i = A_{i||}S_z I_{iz} + A_{i\perp}(I_i^+ S^- + I_i^- S^+), \quad i = 1, 2$$

describe contact interaction of the electron spin with the nuclear spins of the first and second groups of equivalent nuclei. Here $S$ and $I_i$ are operators of electron angular momentum and total angular momentum of $i$-th nuclear group.

Let us introduce the representation of the functions $|S, J_1, L_1, J_2, L_2, \alpha\rangle$, where $S = \pm1/2$ is the quantum number of the electron spin $z$-projection, $J_i$ and $L_i$ — are the quantum numbers of the total angular momentum of the $i$-th nuclear group and its $z$-projection, respectively, $\alpha$ is the set of all other quantum numbers needed for the state to be uniquely specified. In this case, the total angular momentum squares of the $i$-th nuclear group is $I_i^2 = J_i(J_i + 1)$ (in the units $\hbar^2$) and the possible values of $L_i$ are: $L_i = -J_i, 1-J_i, \ldots, J_i-1, J_i$. As has been shown in [3, 4], the number of ways that yields the state with the total angular momentum $J$ by summing $2N$ elementary spins $1/2$ is given by the formula:

$$\Gamma_N(J) = C_{2N}^{N-J} - C_{2N}^{N-J-1} \quad J = 1, \ldots, N.$$

Then, (number of states with given numbers $S, J_1, L_1, J_2, L_2$) = $\Gamma_{N_1}(J_1)\Gamma_{N_2}(J_2)$. Now, the Hamiltonian depends on total angular momenta of the two groups of nuclei and, in the above representation, can be broken down into blocks with specified values of the total angular momenta ($J_1$ and $J_2$) of these groups. The fact that Hamiltonian (3) has no matrix elements between the functions with different values of $\alpha$ can be proven in the same way as in [4]. The initial nuclear state (initial nuclear density matrix) is assumed to correspond to infinite temperature. In this case, the probability of each of the nuclear state is the same and equal to $1/(\text{total number of nuclear states}) = 1/2^{2(N_1+N_2)}$. Note that (dimensions of the block with given $J_1$ and $J_2$) = (number of possible projections of electron spin)$\times$(number of possible projections of $J_1$)$\times$(number of possible projections of $J_2$) = $2(2J_1+1)(2J_2+1)$. 


These blocks are independent and, hence, the dynamic equations for the density matrix can be also broken down into blocks. The greatest possible nuclear polarization for the considered case of two groups of equivalent nuclei is obtained for full nuclear polarization in each of the blocks. In this case, the nuclear density matrix \( \rho_n \) has, in such a block, only one nonzero diagonal element corresponding to projection of the total nuclear spin equal to \( J_1 + J_2 \):

\[
\langle J_1, L_1 = J_1, J_2, L_2 = J_2 | \rho_n | J_1, L_1 = J_1, J_2, L_2 = J_2 \rangle = \frac{(2J_1 + 1)(2J_2 + 1)}{2^{2(N_1 + N_2)}}
\]

The number of such blocks in the nuclear density matrix equals \( \Gamma_{N_1} (J_1) \Gamma_{N_2} (J_2) \). Projection of the total nuclear angular momentum in this state is given by the expression

\[
\langle J_z \rangle_{\text{max}} = \frac{1}{2^{2N_1}} \sum_{J_1=0}^{N_1} \sum_{J_2=0}^{N_2} \Gamma_{N_1} (J_1) \Gamma_{N_2} (J_2) (2J_1 + 1)(2J_2 + 1)(J_1 + J_2) = \frac{2N_1 P(N_1) + 2N_2 P(N_2)}{2^{2N_1}}
\]

Thus, for the maximum nuclear polarization \( P_2 \), in the case of two groups of equivalent nuclei, we obtain:

\[
P_2 = \frac{\langle J_z \rangle_{\text{max}}}{2(N_1 + N_2)} = \xi_1^2 P(N_1) + \xi_2^2 P(N_2) = \xi_1^2 P(\xi_1^2 N) + \xi_2^2 P(\xi_2^2 N)
\]

where \( \xi_i^2 \equiv N_i/(N_1 + N_2) \) - is the fraction of nuclei of the \( i \)-th group and \( N = N_1 + N_2 \) is the total number of pairs of nuclei.

For arbitrary number of groups of the equivalent nuclei \( n \), we can obtain, in a similar way, the following expression for the limiting polarization:

\[
P_n = \sum_{i=1}^{n} \xi_i^2 P(\xi_i^2 N), \quad \sum_{i=1}^{n} \xi_i^2 = 1
\]

In the above treatment, the number of nuclei in each group was supposed to be even. Intuitively, it seems plausible that this assumption is not essential provided that the number of nuclei in a group \( \xi_i^2 N \) is much greater than unity. In this case, we can use the approximate formula (2) for \( P(N) \) and obtain:

\[
P_n \approx \frac{1}{\sqrt{\pi N}} \sum_{i=1}^{n} |\xi_i|
\]

For a given number of groups of equivalent nuclei, the greatest nuclear polarization is achieved when all the groups have the same number of nuclei, i.e., \( \xi_i^2 = 1/n \). In this case,
the limiting polarization, for a given total number of pairs of nuclei, exceeds by a factor of \( \sqrt{n} \) that of the box-model \( 1/\sqrt{\pi N} \). Thus, if, for instance, the real wave function of the electron in the quantum dot is presented by 10 levels, the limiting polarization will increase, in this case, not more than by a factor of \( \sqrt{10} \). In the above calculation, the greatest nuclear polarization arises when the number of groups of equivalent nuclei appears to be equal to the number of nuclear pairs \( N \). In this case, Eq. (12) is not precise because the number of nuclei in a group is not large (two nuclei in each). However the limiting polarization can be now obtained using Eq. (11):

\[
P_N = \frac{3}{8}
\]

which is close to \( 1/\sqrt{\pi} \) obtained by Eq. (12). From the above treatment we see, that the model in which the number of groups of equivalent nuclei is only by a factor of 2 smaller than their total amount, cannot describe the 100% nuclear polarization.

III. DISCUSSION

As was already mentioned above, for the quantum dot consisting of \( 10^4 \) nuclei, the box-model (with the number of groups of equivalent nuclei equal to unity) yields the limiting nuclear polarization \(~ 1\%\). As follows from Eq. (12), in the model compatible with the nuclear polarization of 50% obtained in [7], one has to take into account at least \( n = 50^2 = 2500 \) groups of equivalent nuclei. It looks hopeless to obtain exact solution for this model or to analyze it numerically. The only possible approach to these problems can be based on uncontrollable simplifications of its mathematical solution that imply more or less convincing intuitive considerations.

Interpretation of a real experiments using such approaches appears to be vulnerable in two points: (i) inaccuracy of the model itself (insufficiently large number of groups of equivalent nuclei, neglecting dipole-dipole interaction between nuclear spins, etc.), (ii) uncontrollable errors related to simplification of mathematical solution of the model problem. On the other hand, if the experimentalist has some grounds to believe that the nuclear polarization, in his experiments, does not exceed the limiting value \( P_1 \) for the box-model, then the use of this model (with only one of the two vulnerable points listed above) for the interpretation of these experiments may be quite efficient. The models that are more precise than the box-model include mechanisms providing nuclear polarization higher than \( P_1 \). However, if the nuclear polarization, in the experimental run, does not exceed \( P_1 \) (e.g., under sign-alternating optical orientation or under conditions of strong nuclear relaxation), then these mechanisms can be considered as non-efficient (sign-alternating orientation) or suppressed (strong nuclear relaxation), and the box model can be used to interpret the experimental data.

Keeping in mind all the aforesaid, let us discuss, in a more general sense, applicability of Hamiltonian (1) for description of the experiments on spin dynamics of quantum dots. Within what time intervals can Hamiltonian (1) be used for description of the electron-nuclear spin dynamics in a quantum dot? How many groups of equivalent nuclei should be chosen? The experiments on spin dynamics of a quantum dot show that, for the repetition rate of the pump pulses of about 100 MHz, the electron polarization reaches its steady-state value (\(~ 1/3 \) in zero magnetic field) for the time much shorter than the pulse repetition period. The estimates show that, within this time scale, the internuclear dipole-dipole
spin interaction and the electron-nuclear dipole interaction \([8]\), neglected in Hamiltonian (1), are of no importance. The state of nuclear subsystem cannot substantially change during one period of optical orientation, because the angular momentum transferred to the nuclear subsystem for this time interval cannot exceed 1. For this reason, application of Hamiltonian (1) to calculation of time dynamics of the electron spin between the pump pulses seems justified. However, in this case, along with initial value of the electron spin, one has to specify initial state of the nuclear subsystem. Consider now, in a qualitative way, the nuclear dynamics. If we assume that the nuclear system acquires from electron, with each pump pulse, the angular momentum 1, then the quantum dot consisted of 10^4 nuclei with spins 1/2 will reach the degree of nuclear polarization of 1% not earlier than after 100 pulses. Such a nuclear polarization is compatible with the box-model. This is why, during, at least, the first 100 pulses, the electron-nuclear spin dynamics calculated in the framework of the box-model will hardly strongly differ from that calculated using the model with larger number of groups of equivalent nuclei. Moreover, the studies of exact solution of the box-model allow one to point out the reasons why, in fact, more than 100 pulses are needed to achieve the nuclear polarization of \(\sim 1\%\). The point is that, under periodic orientation of the electron spin in the quantum dot, a regularity arises in the nuclear density matrix \([4]\) leading to quasi-periodic dynamics of the electron spin. The quasi-periodicity reveals itself in the fact that, just before the arrival of the next pump pulse, the electron polarization in the quantum dot increases to the value close to that just after the previous pulse. As a result, the rate of the angular momentum transfer to the nuclear system substantially decreases and the nuclear polarization process slows down. The above arguments provide grounds to conclude that the electron-nuclear spin dynamics of the box-model will not strongly differ from that of the model with larger number of the groups of equivalent nuclei within the time interval essentially exceeding 100 periods of optical orientation.

In summary, we can conclude that: (i) The initial stage of the electron-nuclear spin dynamics (when the nuclear polarization does not exceed \(P_1\)), can be described in a plausible way using the box-model. This stage corresponds to a few first hundreds of the pump pulses. (ii) The refinement of the box-model aimed at description of the nuclear polarization exceeding \(P_1\) by increasing the number of groups of equivalent nuclei \(n\) is characterized, on the one hand, by a slow (square-root) increase of the limiting polarization \(P_n \sim \sqrt{n}P_1\) and, on the other, by impossibility of the exact mathematical analysis.

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