Spin-engineered quantum dots

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Spatially nonhomogeneously spin polarized nuclei are proposed as a new mechanism to monitor electron states in a nanostructure, or as a means to create and, if necessary, reshape such nanostructures in the course of the experiment. We found that a polarization of nuclear spins may lift the spin polarization of the electron states in a nanostructure and, if sufficiently strong, leads to a polarization of the electron spins. Polarized nuclear spins may form an energy landscape capable of binding electrons with energy up to several meV and the localization radius > 100\AA.

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\textbf{I. INTRODUCTION}

Progress in microelectronics depends crucially on a deep understanding of the electronic properties of low dimensional semiconductors and nanostructures. Typical examples are heterojunctions, quantum wells, and fabricated out of them, 1D quantum wires and 0D quantum dots.
Characteristics of nanostructure devices are mainly determined by the shape of the potential landscape and can be monitored by applying a gate voltage and/or an external magnetic field. A possible role of polarized nuclear spins is generally overlooked in this context. The technique of interband optical pumping, developed in the 60th (1) (see also (2)) opened up a way to reach nuclear spin polarization approaching 50%. This results in an effective magnetic field (called Overhauser field) acting on the electron spins which may reach a few Tesla, or equivalently, several meV energy. This energy is comparable to the electron energies in typical nanostructure devices.

As a consequence, one may expect that through a controlled polarization or depolarization of nuclear spins one may monitor the characteristics of a nanodevice. The Overhauser field leads to an effective Zeeman splitting of the electron states and subsequently to their shifts. If this effect is sufficiently strong, it may lead to a polarization of the electrons in the dot.

Since, at low temperature, the characteristic time of nuclear spin relaxation is extremely long, one may think about creating a potential landscape for the electrons by a spatially inhomogeneous polarization of the nuclei. By polarizing nuclei in a small region one can create a local potential, attractive for electrons with one spin orientation and repulsive for electrons with the opposite spin orientation. This may open up a new way of spin-engineering of quantum dots or geometrically more complex nanodevices, whose shapes can be manipulated in real time.

All semiconductor materials consist of more than one stable elemental isotope with non zero nuclear spin \( I \); for example \(^{69}\text{Ga} \) (natural abundance 60.4%), \(^{71}\text{Ga} \) (31.6%), \(^{75}\text{As} \) (100%), all have nuclear spin \( I = \frac{3}{2} \), while \(^{27}\text{Al} \) has \( I = \frac{5}{2} \). A lithographically prepared GaAs/Al\(_{1-x}\)Ga\(_x\)As heterostructure of dimensions 10nm×10nm comprises a huge number of active nuclear spins \( \sim 10^4 \). In (Al)GaAs the amount of active nuclei is of the order of the total number of atoms in that volume. In Si/Si\(_{1-x}\)Ge\(_x\) heterojunctions the active nuclear spins have substantially lower concentration because the natural abundances of the Ge and
Si isotopes with nonzero nuclear spins are rather small (7.6% for $^{73}$Ge with $I = \frac{9}{2}$ and 4.7% for $^{29}$Si with $I = \frac{1}{2}$). However, recent progress in nanotechnology and growth of isotopically controlled bulk Si [3] and Ge [4], superlattices of, e.g., $^{70}$Ge/$^{74}$Ge [5] will allow one to fabricate low dimensional semiconductor structures with controlled abundances of spin active and neutron transmuted nuclei.

Nuclear spin diffusion decreases with the dimensionality [6] of the system. Earlier estimates [7] show that nuclear spin relaxation times in conventional pure semiconductors are very long. At helium temperature they are at least of the order $10^2$ to $10^3$ s and can reach up to a few hours. As a comparison, the electron spin relaxation times are about $10^{-7}$s. It means that any time variation of the nuclear polarization occurs adiabatically slow as compared to the time scales of the electron dynamics. Therefore, one may consider any potential created by the polarized nuclei for the electron subsystem as quasi-static. Vagner et al [8] proposed a new type of Aharonov-Bohm effect caused by spin polarized nuclei in the absence of any magnetic field, which was observed very recently [9]. Ref. [10] discusses an anomalous Hall effect caused by the hyperfine interaction of polarized nuclei and electrons. In a recent paper [11] it was shown that a rather small nuclear spin polarization can contribute to the electron relaxation in a quantum dot.

Due to the enormous difference of nuclear, $m_n$, and electron, $m_e$, masses, the Zeeman splitting is substantially smaller for nuclei as compared to that for electrons. It makes polarization of nuclei by an external magnetic field much more difficult. Nevertheless, various optical techniques [12,13] for polarizing nuclear spins via creating nonequilibrium spin polarized electrons, which transfer their polarization to the nuclear subsystem in the course of thermal equilibration of electrons, lead to much better results. These techniques are much more efficient than the direct magnetization by an external magnetic field and result in much higher nuclear spin polarizations.

Overhauser [14,15] has described the hyperfine interaction of the electron and the nuclear spins in a solid through the Fermi-like contact potential
\[ H_{hf} = A S \sum_i I_i \delta(\mathbf{r} - \mathbf{r}_i) \equiv g \mu_B \hat{\mathbf{B}}_n \cdot \mathbf{S} \quad (1) \]

where the summation in (1) is over the spin active nuclei, \( \mu_B \) is the Bohr magneton, and \( g \) is the electron \( g \)-factor. \( A \) is the coefficient of the hyperfine interaction, which is specific for each particular type of nucleus. The operator \( \hat{\mathbf{B}}_n \) averaged over the nuclei and the electron wave functions results in the hyperfine Overhauser field, \( \mathbf{B}_n \), which acts on the electron spin.

In GaAs the hyperfine coefficient \( A \) is negative and the Overhauser field tends to polarize the electron spins parallel to the nuclear spins. This field may be large [15–19] depending on the type of nuclei and the degree of their polarization. For example, for naturally abundant isotopes in GaAs it reaches the value \( B_n = 5.3 \text{T} \) in the limit that all nuclear spins are completely polarized. The value \( B_n = 1.7 \text{T} \) has been achieved experimentally [2], which corresponds to 32% nuclear polarization. Thus electrons may be strongly influenced by the Overhauser field in a nano-device with polarized nuclear spins, although this field does not manifest itself magnetically due to the smallness of the nuclear magnetic moments.

II. QUANTUM DOT WITH POLARIZED NUCLEAR SPINS.

The aim of this section is to estimate the influence of the Overhauser field, created by a spatially nonhomogeneous nuclear polarization, on the electrons in a quantum dot. We assume that a local Gaussian distribution of nuclear polarization along the \( z \) axis was created,

\[ I_i = I_m \exp \left( -\frac{\rho_i^2}{2a^2} \right) \exp \left( -\frac{z_i^2}{2b^2} \right), \quad (2) \]

where \( \{\rho_i, z_i\} \) are the cylindrical coordinate of the \( i \)-th nucleus. A quantum dot (QD) with spin polarized nuclei is described by the Hamiltonian

\[ H = \left( -\frac{\hbar^2}{2m} \Delta + \frac{m \omega^2 r^2}{2} \right) \delta_{\sigma,\sigma'} + H_{hf}, \quad (3) \]

where for simplicity we assumed a 3D parabolic confined quantum dot with confinement frequency \( \omega \). The QD part in (3) is diagonal with respect to the spin projections \( \sigma \) whereas
$H_{hf}$ is defined by Eq. (1) with the nuclear polarization distribution (2). Here we shall disregard the spin flip processes and consider only the longitudinal part of the hyperfine interaction, i.e., we assume that $S \cdot I_{i} = I_{i} \sigma_{z}$.

The eigenenergies and eigenfunctions of the three dimensional harmonic oscillator representing the quantum dot in the absence of nuclear spin polarization is well known:

$$E_{n_{x}n_{y}n_{z}} = \hbar \omega \left( n_{x} + n_{y} + n_{z} + \frac{3}{2} \right),$$

$$\Psi_{n_{x}n_{y}n_{z}}(\mathbf{r}) = \xi^{3/2} \left( 2^{n_{x}+n_{y}+n_{z}} n_{x}! n_{y}! n_{z}! \sqrt{\pi} \right)^{1/2} \exp \left( -\frac{\xi^{2}(\rho^{2} + z^{2})}{2} \right) H_{n_{x}}(x\xi) H_{n_{y}}(y\xi) H_{n_{z}}(z\xi)$$

where $H_{n}(\ldots)$ is the Hermite polynomials, and $\xi = \sqrt{m\omega/\hbar}$ determines the inverse size of the dot.

Now we calculate the first order perturbation correction to the ground state energy

$$E^{(1)}_{0\sigma} = \frac{\sigma A}{\Omega} I_{m} \frac{2\sqrt{2a^{2}b\xi^{3}}}{[1 + 2a^{2}\xi^{2}]^{1/2}}$$

where $\Omega$ is the volume per nuclear spin, and $\sigma = \pm 1$ is the direction of the electron spin.

The second order correction is due to virtual excitations to even states of the oscillator for the chosen spatial distribution of the nuclear spin polarization. The contribution of the lowest 3-fold degenerate state with the energy $E_{200} = E_{020} = E_{002} = \frac{7}{2}\hbar \omega$ is

$$E^{(2)}_{0} = -\frac{2A^{2}I^{2}_{m}}{\hbar \omega \Omega^{2}} \frac{a^{4}b^{2}\xi^{6}}{[1 + 2a^{2}\xi^{2}][1 + 2b^{2}\xi^{2}]} \left\{ \frac{2}{[1 + 2a^{2}\xi^{2}]^{2}} + \frac{1}{[1 + 2b^{2}\xi^{2}]^{2}} \right\}.$$  

The energy of the ground state second order perturbation theory becomes

$$E_{0\sigma} = \frac{3}{2}\hbar \omega + E^{(1)}_{0\sigma} + E^{(2)}_{0}.$$  

If the nuclei are polarized in a region, exceeding the size of the dot (i.e., $a\xi, b\xi > 1$) then this energy becomes

$$E_{0\sigma} = \frac{3}{2}\hbar \omega + \sigma \frac{AI_{m}}{\Omega} - \frac{A^{2}I^{2}_{m}}{16\Omega^{2}\hbar \omega \xi^{4}} \frac{1}{[a^{4} + b^{4}]}.$$  

We see that that the principal effect of the Overhauser field on the electron levels is the Zeeman-like lift of the spin degeneracy. It holds also for a homogeneous polarization of
the nuclear spins, when $a, b \to \infty$. A nonhomogeneous polarization may shift the electron levels. The ground state (9) is shifted downward regardless of the $\sigma$ value. However, one should keep in mind, that accounting for the nuclear spin contribution, the potential at the bottom of the well becomes now negative, $-A_{m}m_{\Omega}$, meaning that the distance of the ground state from the bottom of the well may, in fact, increase.

According to the estimates, presented above, the Zeeman-like splitting caused by the first order correction (9) linearly depends on the nuclear spin polarization and may become substantial. The condition $A_{m}m_{\Omega} > \hbar \omega$ can be easily achieved. Although under this condition one cannot restrict oneself to the lowest order terms of perturbation theory, nevertheless the general pattern is qualitatively clear, which is illustrated in Fig.1. As an example we consider a quantum dot occupied by six electrons. It is well known that the Pauli principle makes the total spin of a dot with an even number of electrons equal to zero, when the nuclear polarization is zero and the spin degeneracy is not lifted, as is situation shown in Fig. 1(a). At relatively small $A_{m}m_{\Omega} < \hbar \omega$ polarization the spin degeneracy is lifted but the spin configuration of the electrons in the dot does not change, Fig. 1(b). There are still three spin up electrons and three spin down electrons, so that the net spin of the dot remains zero. However, at larger nuclear polarizations, $A_{m}m_{\Omega} > \hbar \omega$, the Zeeman-like splitting becomes larger than the energy distance between the levels, which results in a new spin configuration of the electrons in the dot. One can see from Fig. 1(c) that the level $n = 3$ for spin up electrons moves below the $n = 2$ level for the spin down electrons. As a result one electron flips its spin. We now have four spin up electrons and only two spin down electrons, which results in a net electron spin of the dot equal to 1.
FIG. 1. A graphical illustration of the polarization of a quantum dot with even number of electrons. (a) shows four lowest levels of the dot occupied by six electrons in the absence of the nuclear spin polarization. (b) The nuclear spin polarization causes a Zeeman-like splitting of the levels which is still less than the distance between the levels. No net spin appears in the dot. (c) The Zeeman-like splitting exceeds the distance between the levels resulting in a spin polarized electron state.

III. POTENTIAL CREATED BY POLARIZED NUCLEAR SPINS

Having considered the role played by spin polarized nuclei in a conventional quantum dot, we address now the problem of engineering potential landscapes by means of polarized nuclei. This may be achieved by creating a spatially inhomogeneous nuclear spin polarization. For $A < 0$ the resulting potential (III) is attractive for spin up electrons (parallel to the nuclear spin polarization) and repulsive for the spin down electrons (antiparallel to the nuclear spin polarization). One may create a nuclear spin polarization $I_m$ in a region with a size $a$, thus forming an attractive (for spin up electrons) potential $U_{hf} = \frac{AI_m}{\Omega}$. Then a simple reasoning based the uncertainty principle leads us to the conclusion such a potential may bind an
electron if the condition

\[ a > \hbar \sqrt{\frac{\Omega}{2m^*|A||I_m}}} \]  

(10)

is fulfilled.

An estimate can be made for the case of GaAs. Creating a 30% nuclear spin polarization results in a potential \( U_{hf} \sim 3\text{meV} \) which is capable of binding an electron in a well of typical size \( a > 100\text{Å} \).

IV. CONCLUSIONS

We discussed the possible influence of the hyperfine interaction between spin polarized nuclei and electrons and its influence on the electron subsystem with a special emphasis to confined regions in semiconductors, e.g., quantum dots. An effective Overhauser magnetic field of the polarized nuclei lifts the spin degeneracy and is therefore, capable of polarizing electrons in a quantum dot. Long lived nonhomogeneously spin polarized nuclei may create a local potential, attractive for electrons with one spin direction and repulsive for the electrons with the opposite spin directions. Such a potential can form, e.g., a quantum well, a quantum dot or any other nanostructure, depending on the spatial engineering of the polarized nuclei.

The interesting feature of such potential landscapes is that they can be created and reshaped, if necessary, in real time by polarizing and/or depolarizing nuclear spins. We believe that this sort of technique, being developed experimentally, may open a new promising venue in nanotechnology which we call spin-engineering.

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