Spin relaxation in semiconductor quantum dots

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We have studied spin-flip processes in GaAs electron quantum dots that accompany transitions between different discrete energy levels. Several different mechanisms that originate from spin-orbit coupling are shown to be responsible for such processes. We have evaluated the rates for all mechanisms with and without a magnetic field. We have shown that the spin relaxation of the electrons localized in the dots differs strikingly from that of the delocalized electrons. The most effective spin-flip mechanisms related to the absence of the inversion symmetry appear to be strongly suppressed for localized electrons. This results in unusually low spin-flip rates.

Quantum dots (QD's) are small conductive regions in semiconductor structures that contain a tunable number of carriers. The shape and size of quantum dots can be controlled by the gate voltage. The localized electronic states in QD's can be significantly modified by a magnetic field. All this provides a valuable opportunity to study the properties of the electron quantum states in detail and manipulate the electrons in these artificial atoms in a controllable way (see Refs. 1 and 2 for review).

The spin states in quantum dots are considered to be promising for physical realization of the quantum computation algorithm.[3] Quantum computation requires coherent coupling between the dots, the coherence to be preserved on sufficiently long time scales. That makes it relevant to provide a complete theoretical estimation of the typical spin dephasing time of the electron in the QD. Transport experiments with QD's have revealed that the current through a quantum dot can be influenced by the spin effects.[4] It opens up a possibility to estimate spin relaxation rates by means of transport measurement.[5] The origin of this effect is that the spin-flip process can provide a bottleneck for the energy relaxation in the dot, i.e., for transitions between the excited and ground states. Indeed, in the absence of the spin flip the total spin of the dot is a good quantum number and no transition is allowed between the states of different total spins. To illustrate, let us consider a QD with two electrons which can be placed in two levels. The ground state corresponds to two electrons in the lowest level having opposite spins ($S=0$). One of the excited states corresponds to two electrons in different levels having the same spin direction ($S=1$). Due to the Pauli principle, the electron in the upper level cannot get to the lower level without changing its spin. Therefore, the relaxation to the ground state of the dot has to be accompanied by a spin flip.

In contrast to the situation in two-dimensional (2D) electron gas the electrons confined in the dot experience no electron-electron scattering (see Ref. 6). The only source of dissipation is the interaction with phonons. Moreover, although the electron-electron interaction is quite important in determining the energies of the states and the number of electrons in the dot, it is less important for the structure of the wave functions. To calculate the matrix elements, we approximate the many-electron wave functions by the Slater determinants. In this way, we can treat the spin-flip processes within the one-electron approach. The electron-electron interaction can change only the numerical factors in our results.

Since most controllable QD's are made on the basis of 2D electron gas GaAs heterostructures with the [100] growth direction, we concentrate on the spin-flip mechanisms that are relevant for GaAs, and for this confinement direction. Such mechanisms are very specific for $A_{III}B_V$ compounds. The unit cell has no inversion symmetry, which gives rise to a strong spin-orbit splitting in the electron spectrum. The splitting is known[7,8]to be the main source of the spin-flip both in the 3D and 2D cases. Besides, the piezoelectric effect provides a strong coupling of electrons to the acoustic phonons. Such coupling may be important for the inelastic relaxation in the GaAs crystal both with and without a spin-flip.

The spin relaxation of the delocalized electrons in GaAs 2D electron gas has been thoroughly studied.[8] Some spin-flip mechanisms that are effective in the 3D case, for instance, the Yafet-Elliot mechanism,[7] do not work in two dimensions. The most effective mechanisms in the 2D case are related to the broken inversion symmetry, either in the elementary crystal cell or at the heterointerface.[8,9] Those are described by the terms in the electron Hamiltonian[8] that are linear in the two-dimensional electron momentum and proportional to the first power of the small parameter $\Delta/E_g \ll 1$, Δ being the spin-orbit splitting of the valence band of the bulk GaAs crystal and E_g the band gap. Hence, the spin-flip rate is proportional to $(\Delta/E_g)^2$.[8]

In this paper we show that the zero-dimensional character of the states in the quantum dot leads to further suppression of the spin-flip rate. We reveal a fairly interesting fact that the terms which are linear in Δ/E_g disappear from the transition matrix elements in the true two-dimensional case. The contributions to the spin-flip rate which are quadratic in Δ/E_g appear only if we take into account either the admixture of the higher states of the size quantization in the z direction, i.e., the weak deviation from the true 2D motion, or the higher orders in the expansion over the electron momentum in the plane, $\propto p^3$. Thus, such contributions acquire extra small factors: either the ratio of the lateral kinetic energy E_{lat} to the distance between the quantized levels in the

z-direction E_z, or E_{lat}/E_g. Therefore, they may become comparable to the competing term $\propto(\Delta/E_g)^6$ that stems from the third order perturbations in terms $\propto\Delta/E_g$.

We start with the following one-electron Hamiltonian which is derived from the Kane model (see Ref. 7) and describes the electron in the conduction band in the presence of magnetic field **B** normal to the 2D plane, arbitrary confining potential $U(\mathbf{r})$, both vertical and lateral (possible impurity potential should be also added to U), and the phonons:

$$\hat{\mathcal{H}}=\frac{\hat{\mathbf{p}}^2}{2m}+U(\mathbf{r})+U_{ph}(\mathbf{r},t)+\frac{1}{2}g\mu_B\hat{\sigma}_z B+\sum_{i=1}^{4}\hat{\mathcal{H}}_i;$$

$$\hat{\mathcal{H}}_1=\frac{\hbar\Delta}{3mE_g^2}\hat{\boldsymbol{\sigma}}[\boldsymbol{\nabla}U\cdot\hat{\mathbf{p}}];$$

$$\hat{\mathcal{H}}_2=\frac{2\Delta}{3\sqrt{2mE_g}m_{cv}E_g}\hat{\boldsymbol{\sigma}}\cdot\hat{\boldsymbol{\kappa}};\quad \hat{\mathcal{H}}_3=\frac{1}{2}V_0\hat{\boldsymbol{\sigma}}\cdot\hat{\boldsymbol{\varphi}}; \tag{1}$$

$$\hat{\mathcal{H}}_4=\tilde{g}\mu_B(u_{xz}\hat{\sigma}_x+u_{yz}\hat{\sigma}_y)B$$

Here $\hat{\mathbf{p}}=-i\hbar\boldsymbol{\nabla}+(e/c)\mathbf{A}$ is the 3D electron momentum operator, m the effective mass, $\hat{\boldsymbol{\sigma}}$ the Pauli matrices. The first three terms in the Hamiltonian do not depend on the spin. The third term describes the spin-independent interaction with the phonons, including the piezoelectric ones. Note, that we use the conventional model of 3D phonons. The fourth term is the Zeeman energy. The other four terms describe the spin-orbit effects. $\hat{\mathcal{H}}_1$ is due to the relativistic interaction with the electric field caused by the confinement or impurities. It is enhanced due to the band effects. $\hat{\mathcal{H}}_2$ stems from the absence of the inversion symmetry in the bulk. Here, m_{cv} is the parameter of the Kane model, $\hat{\kappa}_x=\hat{p}_x(\hat{p}_y^2-\hat{p}_z^2)$ and the other components are obtained by the cyclic permutation of the indices, x,y,z being the main crystallographic axes. In the true 2D case the averaging of $\hat{\mathcal{H}}_2$ with the wave function of the first quantized level in the z direction results in the spin splitting proportional to $p_{x,y}$.[8] $\hat{\mathcal{H}}_3$ describes the spin-orbit splitting of the electron spectrum due to the strain field produced by the acoustic phonons. There, $\hat{\varphi}_x=(1/2)\{u_{xy},\hat{p}_y\}_+-(1/2)\{u_{xz},\hat{p}_z\}_+$, where $\{,\}_+$ denotes the anticommutator. The other components are obtained by cyclic permutations, u_{ij} is the lattice strain tensor, and $V_0\propto\Delta/E_g$ is the characteristic velocity whose value is well known for GaAs,[10] $V_0=8\times10^7$ cm/s. In GaAs, the electron g-factor ($g=-0.44$) differs strongly from the free electron value $g_0=2$ owing to the strong spin-orbit interaction, which mixes the valence-band and conduction-band states.[11] The admixture should depend on the lattice deformation, leading to a new mechanism of spin-phonon coupling in GaAs in the presence of an external magnetic field (see also Ref. 12). Coefficient $\tilde{g}$ can be found within the Kane approach, $\tilde{g}=(2m_0/\sqrt{3}m)(\Delta/E_g)(d/E_g)$, d (order of several eV) is one of the three deformation constants describing the strain effect on the hole-band splitting.[7]

The first two terms in Hamiltonian (1) define a series of discrete electron states in the dot. Most of the relations given below hold for the arbitrary states in the general confinement potential. To get down to concrete numbers, we assume either an elliptic or circular dot in the parabolic confinement potential that can be characterized by frequencies ω_x,ω_y, and concentrate on the transitions between the low-lying states. Such transitions occur with phonon emission. The most probable process would be that without a spin flip. As we mentioned above, we assume that this process is forbidden by the Pauli principle. Therefore, we calculate the rates of the spin-flip processes with phonon emission for different mechanisms related to spin-orbit terms $\hat{\mathcal{H}}_{1-4}$ both with and without a magnetic field. Since the spin-orbit coupling for electrons is relatively weak, we treat these terms in the framework of the perturbation theory. The rates obtained depend on the energy difference between the states.

The four spin-orbit terms in Eq. (1) generate a variety of different spin-flip mechanisms. They can be subdivided into two groups. The mechanisms of the first group are due to the spin-orbit admixture of different spin states. In the presence of spin-orbit terms $\hat{\mathcal{H}}_1,\hat{\mathcal{H}}_2$ the electron spin-up state actually acquires a small admixture of the spin-down state. This leads to a non-vanishing matrix element of the phonon-assisted transition between two states with opposite spins. The phonon itself does not flip the spin and provides only energy conservation here. We revealed that $\hat{\mathcal{H}}_2$ always provides bigger admixture than $\hat{\mathcal{H}}_1$. In particular, it is possible to show that spin-orbit coupling with the lateral electric field gives rise to a weaker effect. However, as we mentioned above, the effect of the admixture on the matrix element tends to disappear in the first order of the perturbation theory. The effect persists in (i) the third order of the perturbation theory and appears in the first order if we take into account (ii) the third-order terms in the lateral momentum; (iii) the admixture of the higher states of the size quantization in the z direction; (iv) the impurity potential; (v) the Zeeman splitting in the magnetic field. Thus, in this group we have five mechanisms to compare. The mechanisms of the second group are due to direct spin-phonon coupling. They are described either by $\hat{\mathcal{H}}_3$ or $\hat{\mathcal{H}}_4$.

It is not clear *a priori* which mechanism is the most effective. Below, we consider the mechanisms one by one.

Admixture mechanisms. We begin with the derivation of the general expression, which gives the phonon-assisted transition rate between states 1 and 2 when there is no direct coupling between the spin and the phonon. The wave function of each state is a two-component spinor, $\Psi^i(\mathbf{r})$, $i=\uparrow,\downarrow$. In the absence of direct spin-phonon coupling only the scalar product of the two spinors, $\Psi_1^{\dagger i}(\mathbf{r})\Psi_2^i(\mathbf{r})$, enters the matrix element. We take into account only the coupling to the piezo-phonons which is known to be the most effective one in polar crystals for energy transfer ϵ less than 10 K.[13] It is characterized by the piezomodulus h_{14}, $eh_{14}=1.2\times10^7$ eV/cm for GaAs. As explained below, we take into account only transverse phonons and obtain

$$\Gamma_{12}=\frac{2\pi}{\hbar}2\frac{\hbar(eh_{14})^2}{2\rho s_t}\int\frac{d^3Q}{(2\pi)^3}\frac{A_t(\mathbf{Q})}{Q}\Bigg|\times\int d^3r\exp(i\mathbf{Qr})\Psi_1^{\dagger i}(\mathbf{r})\Psi_2^i(\mathbf{r})\Bigg|^2\delta(\hbar s_t Q-\epsilon), \tag{2}$$

where $\mathbf{Q}$ is the phonon wave vector, s_t the transverse sound velocity, ρ the crystal mass density, and $A_t(\mathbf{Q})$ the anisotropy factor for the transverse phonon that depends on the orientation of $\mathbf{Q}$ with respect to the main crystallographic axes. We consider the transitions between the neighboring low-lying discrete energy levels, so that $\epsilon=\hbar sQ\approx\hbar^2/m\lambda^2$, λ being the typical dot size in the lateral direction. Since the energy transfer is of the order of the electron energy itself, and the perturbation theory in phonons requires that this energy exceeds ms^2, from the condition $\epsilon\gg ms^2$ we obtain $Q_z\gg q\approx 1/\lambda$. This means that the phonon is emitted almost perpendicular to the 2D plane. Then, assuming that $Q_z z_0<1$, z_0 being the width of the 2D layer in the z- direction, we can easily calculate the integral over Q_z and over $\mathbf{q}=(Q_x,Q_y)$. The anisotropy factor for the [100] orientation becomes[14] $A_t(\mathbf{Q})\approx 2q^2/Q_z^2\ll 1$, the longitudinal phonons give a much smaller contribution, and Eq. (2) reduces to

$$\Gamma_{12}=-\frac{2s_t\hbar^2(eh_{14})^2}{\rho\epsilon^3}\int dxdyK_{12}^{\star}(\nabla_x^2+\nabla_y^2)K_{12}, \quad (3)$$

where we decompose the wave functions into the lateral and transverse parts, $\Psi(\mathbf{r})=\Phi(x,y)\chi_0(z)$; $K_{12}(x,y)\equiv\Phi_1^{\dagger i}\Phi_2^i$. In the absence of the spin-orbit interaction, $K_{12}\neq 0$ only if states 1,2 have the same spin. The spin-orbit interaction mixes up the spinor components resulting in nonzero K_{12} even if 1,2 have opposite spins.

As a reference, we give the expression for the transition rate *without* a spin-flip which follows from Eq. (3) for the transition $n_x=1$, $n_y=0\Rightarrow n_x=0$, $n_y=0$ in the elliptic quantum dot ($\epsilon=\hbar\omega_x$)

$$\Gamma_{\uparrow\uparrow}=\Gamma_{\downarrow\downarrow}=\frac{\Gamma_0}{4}\sqrt{\frac{\omega_y}{\omega_x}}\left(3+\frac{\omega_y}{\omega_x}\right);\quad \Gamma_0=\frac{2(eh_{14})^2m^2s_t}{\pi\rho\hbar^2\epsilon}. \quad (4)$$

Γ_0 has the value of 3.1×10^8 s^{-1} for the transfer energy of 1 K. Let us consider the spin-flip transitions. The projection of Eq. (1) to the lateral wave functions yeilds the following 2D spin-orbit Hamiltonian:

$$\hat{\tilde{\mathcal{H}}}_2=\beta(-\sigma_x\hat{p}_x+\sigma_y\hat{p}_y);\quad \beta=\frac{2}{3}\langle p_z^2\rangle\frac{\Delta}{(2mE_g)^{1/2}m_{cv}E_g}. \quad (5)$$

Constant β in Eq. (5) depends on the confinement strength and takes the values in the interval $(1\div 3)\cdot 10^5$ cm/s for GaAs heterostructures. Term $\hat{\tilde{\mathcal{H}}}_2$ leads to a nonzero value of K_{12}. At first sight, K_{12} should be proportional to the first power of β. However, in contrast to the extended 2D states, in quantum dots we can actually remove the terms linear in β from the Hamiltonian by the following spin-dependent unitary transformation:

$$\Phi=\left[\hat{I}+\frac{im\beta}{\hbar}(x\hat{\sigma}_x-y\hat{\sigma}_y)+O(\beta^2)\right]\Phi'. \quad (6)$$

We stress that the boundedness of the electron wave functions is essential to this procedure. The spin-dependent term $\propto\beta^2$ in the resulting Hamiltonian contains only $\hat{\sigma}_z$ so that it causes no spin-flip. The terms $\propto\beta^3$ should be taken into account, which gives the rate $\propto\beta^6$. Here we give the result for the case of a circular dot ($\omega_x=\omega_y=\omega_0$) for the phonon-assisted spin-flip transition between the first excited and ground states ($\epsilon=\hbar\omega_0$):

$$\Gamma_1=\frac{8}{3}\Gamma_0\left(\frac{m\beta^2}{\epsilon}\right)^3. \quad (7)$$

The applicability of the perturbation theory in $\hat{\tilde{\mathcal{H}}}_2$ requires $m\beta^2\ll\hbar\omega_0$, so that $\Gamma_1\ll\Gamma_0$, as expected. However, the spin-flip rate exhibits a sharp dependence on β and the lateral confinement energy. For $\beta=1\times 10^5$ cm/s and $\epsilon=10$ K $\Gamma_1\approx 10^{-2}$ s^{-1}, whereas for $\beta=3\times 10^5$ cm/s and $\epsilon=1K$ $\Gamma_1\approx 4.6\times 10^4$ s^{-1}.

Note that the term $\propto\hat{\sigma}_x p_x p_y^2-\hat{\sigma}_y p_y p_x^2$ in Hamiltonian $\hat{\mathcal{H}}_2$ [Eq. (1)] cannot be removed by the above mentioned transformation and gives contribution to the value of K_{12} in the first order of the perturbation theory. The result for the transition $n_x=1$, $n_y=0\Rightarrow n_x=0$, $n_y=0$ in the elliptic quantum dot ($\epsilon=\hbar\omega_x$) reads

$$\Gamma_2=\frac{3}{4}\frac{m\beta^2\epsilon}{E_z^2}\Gamma_0\sqrt{\frac{\omega_y}{\omega_x}}\left[\left(1+\frac{\omega_x}{\omega_y}\right)\left(\frac{\omega_x+\omega_y}{2\omega_x+\omega_y}\right)^2+\left(1+5\cdot\frac{\omega_y}{\omega_x}\right)\frac{\omega_y^4}{(\omega_x^2-4\omega_y^2)^2}\right], \quad (8)$$

where $E_z=\langle p_z^2\rangle/m$. This contribution does not depend on ϵ and E_z and equals $12s^{-1}$ for a circular dot. Since rate Γ_1 falls off with increasing energy, contribution Γ_2 prevails at $\epsilon\gg\sqrt{m\beta^2E_z}$. This crossover energy ranges from 1 to 6 K when β ranges from 1 to 3×10^5 cm/s.

Besides, there are contributions to the spin-flip rate proportional to β^2, which are related either to the virtual transitions to the higher quantized energy levels in the z direction or to the presence of an impurity potential that leads to nonseparability of the transverse (z) and longitudinal variables in Hamiltonian (1). It can be proven that both effects give small contributions to the rate not exceeding Γ_2. Virtual transitions yield the rate $\Gamma_3\simeq\Gamma_2\min(1,\epsilon^2/ms^2E_z)$ which is $\simeq 1$ s^{-1} at $\epsilon=1$ K. For impurities we estimate $\Gamma_4\simeq\Gamma_2(U_{imp}/\epsilon)^2(z_0/r_c)^2$, z_0 being the thickness of the 2D layer in the z direction, $r_c\gg z_0$ the correlation radius of the donor potential, and U_{imp} the magnitude of the donor potential fluctuations. We assume that the latter does not exceed $E_{lat}\simeq\epsilon$, so that $\Gamma_4\ll\Gamma_2$.

The finite Zeeman splitting in the energy spectrum also leads to contributions $\propto\beta^2$. Here we give the final expression for the case of a circular dot ($\omega_x=\omega_y=\omega_0$) in the arbitrary magnetic field $\omega_c\simeq\omega_0$ ($\omega_c=eB/mc$). The solutions of the unperturbed Hamiltonian are the Darwin-Fock states characterized by two quantum numbers: n,l. We consider transitions $n=0$, $l=\pm 1$, $\uparrow\Rightarrow n=0$, $l=0$, $\downarrow$ and using Eqs. (3) and (5) obtain:

$$\Gamma_5=6\Gamma_0(B)\left(\frac{m\beta^2\omega}{\hbar\omega_0^2}\right)\left(\frac{g\mu_BB}{\hbar\omega_0}\right)^2,$$
$$\Gamma_0(B)=\frac{2(eh_{14})^2m^2s_t\omega^2}{\pi\rho\epsilon^3}, \quad (9)$$

where the energy transfer $\epsilon(l=\pm 1)=\hbar\omega+\hbar\omega_c l/2$, $\omega=\sqrt{\omega_0^2+(\omega_c^2/4)}$ and $\Gamma_0(B)$ is the inelastic rate without a spin flip. To estimate, we choose $\omega_c\simeq\omega_0\simeq\epsilon$ so that $\Gamma_5\simeq\Gamma_0(m\beta^2/\epsilon)(g\mu_B B/\epsilon)^2$. This contribution has the same energy dependence as Γ_1. It increases with magnetic field approaching Γ_1 at relatively low magnetic fields $g\mu_B B\simeq m\beta^2$ ($B\simeq 0.01\div 0.1$ T). Note that our consideration corresponds directly to a transition between singlet and triplet states for two electrons in vertical QD.[15] Here, depending on the magnetic field value the energy transfer (the distance betwen the two levels) can vary in a wide interval, so that the $\Gamma_0(B)$ value also changes by many orders of magnitude. Here we give an estimation for $B=2.8$ T where $\hbar\omega_c=\hbar\omega_0=5$ meV. At this point the experimental value of $\epsilon\approx 10$ K and $\Gamma_0(B)\approx 1.3\times 10^{+9}$ s^{-1}. Then using $\beta=3\times 10^5$ cm/s we obtain $\Gamma_5\approx 8.4\times 10^{-7}\Gamma_0(B)\approx 1.1\times 10^{+3}$ s^{-1}.

Direct spin-phonon coupling. Term $\hat{\mathcal{H}}_3$ has been employed by D. Frenkel to describe the spin relaxation of the localized states.[16] Adopting his method to our situation and making use of the condition $Q_{x,y}\ll Q_z$ we obtain a general relation

$$\Gamma_6=-\frac{V_0^2\hbar^2}{128\rho s\epsilon}\int dxdy I_{12}^{\star}(\nabla_x^2+\nabla_y^2)I_{12};$$
$$I_{12}\equiv\Phi_1(\nabla_x-i\nabla_y)\Phi_2^{\star}-\Phi_2^{\star}(\nabla_x-i\nabla_y)\Phi_1. \tag{10}$$

Here, we give the final expression for the elliptic dot and the transition $n_x=1$, $n_y=0\Rightarrow n_x=0$, $n_y=0$ ($\epsilon=\hbar\omega_x$)

$$\Gamma_6=\frac{m^3V_0^2\epsilon^2}{128\pi\rho s\hbar^4}\sqrt{\frac{\omega_y}{\omega_x}}\left(1+\frac{\omega_y}{\omega_x}\right). \tag{11}$$

The corresponding rate is found to be very small ($10^{-1}\div 10^{+1}$ s^{-1}) for the energy transfer $1\div 10$ K.

The last contribution comes from $\hat{\mathcal{H}}_4$. The estimation gives $\Gamma_7\simeq(\tilde{g}\mu_B B)^2(m\epsilon^2/\rho s^3\hbar^4)$ or, in other terms, $\Gamma_7\simeq\Gamma_6(\tilde{g}\mu_B B/mV_0 s)^2$. Therefore, Γ_7 exceeds Γ_6 in a relatively strong magnetic field $\simeq 3$ T.

In conclusion, we have calculated the rates for the phonon-assisted spin-flip transitions in a quantum dot for all possible spin-orbit mechanisms. The localized character of the electron wave functions suppresses the most effective intrinsic spin-flip mechanisms related to the absence of inversion symmetry in GaAs-like crystals. The admixture mechanisms clearly dominate. The third-order mechanism [Eq. (7)] dominates in zero magnetic field exhibiting a sharp dependence on lateral energy. The Zeeman splitting mechanism [Eq. (9)] takes over already in relatively low-magnetic fields. If the corresponding rates become very low ($\simeq 10$ s^{-1}), the p^3 mechanism [Eq. (8)] prevails.

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