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Spin relaxation in the impurity band of a semiconductor in the external magnetic field

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Spin relaxation in the impurity band of a 2D semiconductor with spin-split spectrum and hyperfine interaction in the external magnetic field is considered. Two contributions to the spin relaxation are shown to be relevant: the one given by optimal impurity configurations with the hop-waiting time inversely proportional to the external magnetic field and another one related to electron motion over large distances. The average spin relaxation rate is calculated.

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Spin dynamics in semiconductors has attracted much attention in the last decades [1, 2]. In particular, a number of experimental [3 – 9] and theoretical [10 – 15] works are devoted to the investigation of spin relaxation in the impurity band of a semiconductor. An increasing interest to this problem is motivated by experimental observation of up-to-microsecond spin lifetimes in n-doped bulk GaAs and GaAs/AlGaAs heterostructures, which makes them good candidates for the use in possible spintronics applications.

Spin relaxation in the impurity band is usually driven by hyperfine interaction or spin-orbit coupling. Since the nuclear spin relaxation time is very long ($\tau_N \sim$ ~ 0.1 ms), hyperfine interaction can be treated as a random-in-space static magnetic field with the associated spin precession frequency $\omega_N = A/\sqrt{N}$, where A is the hyperfine coupling constant and N is the number of nuclei within the volume occupied by the wave function [16] (the directions of the random magnetic field for electrons located on different impurities are not correlated). For spin-orbit coupling, the associated spin precession frequency $\boldsymbol{\omega}_{\mathbf{p}}$ is a power function of the electron momentum $\mathbf{p}$ [17 – 19] (in the 2D case, $\boldsymbol{\omega}_{\mathbf{p}}$ is linear in $\mathbf{p}$ [19]). As a result, spin-orbit coupling leads to spin rotation in the process of phonon-assisted hops from one impurity to another by the angle $\boldsymbol{\phi} \approx \boldsymbol{\omega}_{\mathbf{p}_0} \Delta r/v_0$, where Δr is the distance between impurities and $\mathbf{p}_0 = m\mathbf{v}_0$ is the under-the-barrier momentum.

There can be several mechanisms of spin relaxation in the impurity band. Like in quantum dots (QDs), spin relaxation might be driven by phonon-assisted transitions between Zeeman sublevels of the ground state of an impurity. This mechanism of spin relaxation is well studied in QDs [20, 21]. For isolated shallow donor or small QD, it gives spin relaxation times of the order of 1s at fields $B \approx 1$T (Zeeman energy $\mathcal{E}_Z \approx 0.3$K) [22, 23]. Other mechanisms of spin relaxation involve electron hops from one donor to another. For such mechanisms, the spin relaxation rate can be roughly estimated as [7, 10, 24]:

$$1/\tau_S = \omega_N^2 \tau_{hc}, \quad 1/\tau_S = \phi^2/\tau_{hc}, \tag{1}$$

for the case of hyperfine interaction and spin-orbit coupling respectively (here τ_{hc} is the characteristic hop waiting time). These equations are based on the classical picture of the angular spin diffusion in a random magnetic field (in the case of hyperfine interaction the direction of spin precession changes randomly after each hop; in the case of spin-orbit coupling the spin rotates in a random direction in the process of a hop). However, this picture does not account for the exponential variation of the hop waiting times:

$$\tau_{h1} = \tau_0 \exp\left(2\Delta r/a\right), \tag{2}$$

$$\tau_{h2} = \tau_0 \exp\left(2\Delta r/a + \Delta\mathcal{E}/T\right) \tag{3}$$

for phonon emission and absorbtion respectively (here Δr is the distance between impurities, $\Delta\mathcal{E}$ is the distance between their energy levels, $a = \epsilon\hbar^2/2me^2$ is the Bohr radius, and T is the temperature). The main consequence of such inhomogeneity is that it is impossible to introduce an universal time scale for the system under consideration. This fact is confirmed by about ten-fold decrease of the experimentally measured spin correlation time in the bulk GaAs at the crossover from hyperfine-interaction-induced to spin-orbit-induced spin relaxation (see Fig.3 in Ref. [7]). The effects of the inhomogeneity on spin relaxation in the absence of the external magnetic field were considered in Refs. [13, 14] for the systems with spin-split spectrum. In particular,

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it was found that there are two essentially different contributions to spin relaxation: the one related to electron hops over the pairs of impurities with the size of the order of the Bohr radius and another one related to the motion over large distances. In Ref. [9], the dependence of the spin relaxation rate on the external magnetic field was measured experimentally in the impurity band of the bulk GaAs. It was found that the relaxation time first increases, then decreases, then increases again as a function of the magnetic field (see Fig.3 in Ref. [9]).

In this letter, we calculate the average spin relaxation rate for the mechanisms described above in the presence of the external magnetic field B. The use of the averaged relaxation rate is justified when relaxation is slow enough so that an electron can walk over a large distance during the spin relaxation time τ_S (in the opposite case the spin relaxation is governed by escape from the regions with slow relaxation to the regions with fast relaxation [14]). The corresponding condition is [14] $\tau_S \gg \tau_C$ (here $\tau_C = \tau_0 \exp(C\xi_0)$ is the hop waiting time for so-called critical bond [25], $\xi_0 = \sqrt[3]{4L_d^2 W/a^2 T}$, C is the coefficient of the order of unity, $W = e^2/\epsilon L_d$ is the width of the impurity band, and $L_d = n_d^{-1/2}$ is the average distance between impurities). We also assume that spin precession in the external magnetic fields is sufficiently fast $\Omega_0 \tau_S \gg 1$ (here Ω_0 is the spin precession frequency in the external magnetic field). In this case, the components of the spin perpendicular to the magnetic field are suppressed due to fast precession, and hereafter they will be neglected. Finally, we assume that the temperature is sufficiently small so that we can neglect activation to the conduction band, assume that $\hbar\Omega_0 \ll T$, and neglect electron-electron interaction.

Our main point is that over a wide range of parameters the main contribution to the spin relaxation is given by the pairs of impurities with the hop waiting time:

$$\tau_{hc} \approx 1/\Omega_0. \tag{4}$$

Indeed, a common feature of the relaxation mechanisms based on the angular spin diffusion in a random magnetic field is that they are suppressed by applying a longitudinal magnetic field with the associated spin precession frequency larger than the inverse correlation time of the random magnetic field. In the simplest case of a pair of impurities with the hop waiting times $\tau_{h1} = \tau_{h2} = \tau_h$ ($\Delta\mathcal{E} \ll T$), the spin relaxation rate is proportional to $\Delta\Omega^2 \tau_h / \left(1 + \Omega_0^2 \tau_h^2\right)$, where $\Delta\Omega$ is the spin precession frequency in the random magnetic field (in the case of hyperfine interaction $\Delta\Omega \approx \omega_N$; in the case of spin-orbit coupling $\Delta\Omega \approx \Omega_0 \phi$, as shown below). The contribution of the pairs to the spin relaxation rate increases exponentially with Δr for $\tau_h < 1/\Omega_0$ and decrease for $\tau_h > 1/\Omega_0$. Taking into account Eqs. (1) and (4), one can estimate the spin relaxation rate on the pairs of impurities as:

$$1/\tau_S = \nu\omega_N^2/\Omega_0, \quad 1/\tau_S = \nu\phi^2\Omega_0, \tag{5}$$

for the case of hyperfine interaction and spin-orbit coupling respectively (here $\nu \approx (a/L_d)^2 T/W$ is the share of the optimal pairs). At sufficiently small magnetic fields the relaxation is due to electron motion over large distances.

Let us proceed to the rigorous formulation of the problem. We start with the system with spin-split spectrum. The Hamiltonian of the system is

$$\hat{H}_0 = \frac{\mathbf{p}^2}{2m} + U(\mathbf{r}) + \hbar\boldsymbol{\sigma}\boldsymbol{\Omega}_0/2 + \hbar\boldsymbol{\sigma}\hat{\alpha}\mathbf{p}/2mL_S, \tag{6}$$

where $U(\mathbf{r})$ is the impurity potential, L_S is the length characterizing the strength of the spin-orbit coupling, $\hat{\alpha}$ is the dimensionless tensor with the components of the order of unity, and $\boldsymbol{\sigma}$ is the vector of Pauli matrices. The last term on the right-hand side is the combination of the Bychkov-Rashba spin-orbit coupling [17] and Dresselhaus spin-orbit coupling averaged over the electron motion in the direction perpendicular to the quantum well [18, 19]. For the following consideration it is convenient to make a transformation, which cancels spin-orbit coupling to the first order in parameters $1/L_S$ and Ω_0 [26, 27]:

$$\hat{H}' = e^{i\boldsymbol{\sigma}\hat{\alpha}\mathbf{r}/2L_S}\hat{H}e^{-i\boldsymbol{\sigma}\hat{\alpha}\mathbf{r}/2L_S}. \tag{7}$$

As a result,

$$\hat{H}_0' = \frac{\mathbf{p}^2}{2m} + U(\mathbf{r}) + \hbar\boldsymbol{\Omega}_0\boldsymbol{\sigma}/2 + \hbar\left[\boldsymbol{\Omega}_0 \times \hat{\alpha}\mathbf{r}/L_S\right]\boldsymbol{\sigma}/2. \tag{8}$$

Let us consider spin relaxation on a pair of impurities caused by spin precession in the random magnetic field. From the Hamiltonian (8) one can derive an equation, describing spin dynamics:

$$\partial\mathbf{S}/\partial t = \left[\left(\boldsymbol{\Omega}_0 + \Delta\boldsymbol{\Omega}(t)\right) \times \mathbf{S}\right], \tag{9}$$

where $\Delta\boldsymbol{\Omega}(t) = \left[\boldsymbol{\Omega}_0 \times \hat{\alpha}\mathbf{r}(t)/L_S\right]$ and the position of an electron $\mathbf{r}(t)$ takes two values: $\mathbf{r}_1$ and $\mathbf{r}_2$ (here $\mathbf{r}_{1,2}$ are the positions of the impurities). To find the random field correlator $\kappa(t) = \langle\Delta\boldsymbol{\Omega}(t)\,\Delta\boldsymbol{\Omega}(0)\rangle$, one needs to calculate the Green function $G_{ij}(t)$ of the kinetic equation for an electron on a pair of impurities:

$$dn_1/dt = -dn_2/dt = n_2/\tau_{h2} - n_1/\tau_{h1}, \tag{10}$$

where $n_{1,2}$ are the probabilities to find an electron at impurity 1 and 2 respectively. Using the Green function of this kinetic equation, we get:

$$\kappa(t) = \sum_{i,j=1,2} \Delta\mathbf{\Omega}(\mathbf{r}_i)\, G_{ij}(\mathbf{r}_i)\, \mathbf{\Omega}(\mathbf{r}_j)\, n_{j0} = \\ = \frac{\Delta\Omega^2}{4\cosh^2(\Delta\mathcal{E}/2T)} \exp(-t/\tau_h), \qquad (11)$$

where $\Delta\mathbf{\Omega} = [\mathbf{\Omega}_0 \times \hat{\alpha}\Delta\mathbf{r}/L_S]$, $\Delta\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ is the size of the pair, $1/\tau_h = 1/\tau_{h1} + 1/\tau_{h2}$, and $n_{i0} = \tau_{hi}/(\tau_{h1} + \tau_{h2})$ is the equilibrium probability to find an electron at impurity i ($i = 1, 2$). Treating the term proportional to $\Delta\mathbf{\Omega}(t)$ in Eq. (9) as a perturbation and using Eq. (11), we get the following evolution equation for the component of the spin parallel to the external magnetic field:

$$\partial S_\parallel/\partial t = -\int \kappa(t') \cos(\Omega_0 t')\, S_\parallel(t - t')\, dt'. \qquad (12)$$

The spin relaxation rate on a pair of impurities is

$$1/\tau_S(\Delta\mathbf{r}, \Delta\mathcal{E}) = \frac{\Delta\Omega^2}{4\cosh^2(\Delta\mathcal{E}/2T)} \frac{\tau_h}{1 + \Omega_0^2\tau_h^2}. \qquad (13)$$

Depending on the strength of the external magnetic field, several regimes can be realized. In the case $\Omega_0 < 1/\tau_0$, the main contribution to the spin relaxation comes from the pairs of impurities with $|\Delta\mathcal{E}| \le T$ and $\tau_h \approx 1/\Omega_0$ (as follows from Eq. (13), this contribution is proportional to the first power of the external magnetic field). The average spin relaxation rate is

$$1/\tau_S = \int 1/\tau_S(\Delta\mathbf{r}, \Delta\mathcal{E})\, d\Delta\mathbf{r} d\Delta\mathcal{E}/W L_d^2. \qquad (14)$$

Substituting Eq. (13) into Eq. (14), we get:

$$\frac{1}{\tau_S} = \frac{\pi^2}{32}\Omega_0 \frac{T}{W} \left(\frac{a}{L_S}\frac{a}{L_d}\right)^2 \ln^3\left(\frac{1}{\Omega_0\tau_0}\right) g(\mathbf{e}_0). \qquad (15)$$

Let us estimate the spin relaxation rate for GaAs-based quantum well. In this case, Bohr radius $a \approx 50$ Å, Bohr energy $\mathcal{E}_0 \approx 200$ K, and we assume that spin relaxation length $L_S = 1000$ Å, donor concentration $n_d = 10^{11}\,\mathrm{cm}^{-3}$, magnetic field $B = 1$T, and $\ln(1/\Omega_0\tau_0) \approx 3$. In this case, $1/\tau_S \approx 7 \cdot 10^5 \mathrm{s}^{-1}$.

In the case $\Omega_0 > 1/\tau_0$, we can neglect the unity in the denominator of Eq. (13). As a result,

$$1/\tau_S(\Delta\mathbf{r}, \Delta\mathcal{E}) = \frac{1}{\tau_0} \frac{[\mathbf{e}_0 \times \hat{\alpha}\Delta\mathbf{r}/L_S]^2 \exp(-2\Delta r/a)}{1 + \exp(\Delta\mathcal{E}/T)}. \qquad (16)$$

In this case, the main contribution to spin relaxation is from the pairs with $\Delta r \le a$ and $|\Delta\mathcal{E}| \le T$. Substituting Eq. (16) into Eq. (14), we get:

$$\frac{1}{\tau_S} = \frac{3\pi\ln 2}{4} \frac{1}{\tau_0} \frac{T}{W} \left(\frac{a}{L_S}\frac{a}{L_d}\right)^2 g(\mathbf{e}_0). \qquad (17)$$

Thus, the spin relaxation rate is saturated at large Ω_0.

In deriving Eq. (15), we assumed that optimal pairs are effectively separated from the rest of the system, i.e. that an electron makes many hops over a pair before leaving it. For this assumption to be valid, it is required that $\Omega_0\tau_C \gg 1$. In the opposite case, electron motion over large distances gives the main contribution to spin relaxation. In this case the spin relaxation rate is proportional to the electron diffusion coefficient [13, 14]:

$$1/\tau_S \sim D \sim 1/\tau_C. \qquad (18)$$

The influence of the external magnetic field on the diffusion coefficient is well known [25]. At low fields $R_C \gg a\xi_0$ (here $R_C = \hbar c/eBa$ is the cyclotron radius and $a\xi_0$ is the optimal hopping length [25]), it can be described in terms of bending of the tunnelling electron trajectory by the external magnetic field perpendicular to the quantum well, which effectively increases the distance between impurities:

$$aC\xi_0 \to aC\xi_0'(H) = aC\xi_0 \left[1 + \frac{1}{240}\left(\frac{aC\xi_0}{R_C}\right)^2\right]. \qquad (19)$$

As a result:

$$1/\tau_S(H) = (1/\tau_S) \exp\left[-\frac{1}{240}\left(\frac{a}{R_C}\right)^2 (C\xi_0)^3\right]. \qquad (20)$$

For GaAs, the exponent in Eq. (20) can be estimated as $0.04C^3 (B/1\mathrm{T})^2 (1\mathrm{K}/T) \left(10^{11}\mathrm{cm}^{-2}/n_d\right)^{1/2}$.

In the regime described by Eqs. (15) and (17), bending of the tunnelling electron trajectory leads to the decrease of the optimal pair size and, consequently, to the decrease of the probability to find such a pair, while the relaxation rate on an optimal pair remains unchanged. As a result, τ_S does not change much due to bending of the electron trajectory as long as R_C is larger than the optimal pair size $R_C \gg a$.

Next, let us consider the spin relaxation caused by hyperfine interaction. In this case, Eq. (13) still can be used with the replacement $\Delta\Omega \to \Delta\Omega' = (4/3)^{1/2}\omega_N$. Using Eq. (13) and following the same procedure as before, we get:

$$\frac{1}{\tau_S} = \frac{\pi^2}{3} \frac{A^2}{N\Omega_0} \frac{T}{W} \left(\frac{a}{L_d}\right)^2 \ln\left(\frac{1}{\Omega_0\tau_0}\right), \qquad (21)$$

$$\frac{1}{\tau_S} = \frac{4\pi\ln 2}{3} \frac{A^2}{N\Omega_0^2\tau_0} \frac{T}{W} \left(\frac{a}{L_d}\right)^2 \qquad (22)$$

for the case $\Omega_0 < 1/\tau_0$ and $\Omega_0 > 1/\tau_0$ respectively. In GaAs, $A \approx 2\cdot 10^{11}\mathrm{s}^{-1}$, $N \approx 500N_L$, N_L is the number of atomic layers in the quantum well, and other parameters are given after Eq. (15). Substituting these parameters into Eq. (21) and assuming that $N_L = 3$, we get the following estimate: $1/\tau_S \approx 5\cdot 10^6\mathrm{s}^{-1}$.

In the case of small magnetic fields $\Omega_0\tau_C \ll 1$ the spin relaxation rate can be estimated as

$$1/\tau_S = \int \left(A^2 t/N\right) dP\left(t\right), \qquad (23)$$

where $P\left(t\right)$ is the probability for an electron to spend time t at an impurity. This probability can be replaced with the probability that an impurity is separated from the rest of the system by the distance $(a/2)\ln\left(t/\tau_0\right)$ in the coordinate space and $T\ln\left(t/\tau_0\right)$ in the energy space. Assuming that the form of the surrounding empty area is given by $2\Delta r/a + \Delta\mathcal{E}/T \leq \ln[t/\tau_0]$, we find the size of the empty area to be $V = -\ln^3\left[t/\tau_0\right] Ta^2 2\pi/6$, and:

$$P\left(t\right) = e^{-V/WL_d^2} = \exp\left(-\ln^3\left[t/\tau_0\right] 2\pi/3\xi_0^3\right). \qquad (24)$$

Substituting Eq. (24) into Eq. (23) and using saddle-point approximation, we get:

$$1/\tau_S = (\pi/2)^{1/4}\left(2A^2\tau_0/3N\right)\xi_0^{3/4}\exp\left(\frac{2}{3}\sqrt{\xi_0^3/2\pi}\right). \qquad (25)$$

The influence of the external magnetic field on the orbital motion can be accounted for by the replacement:

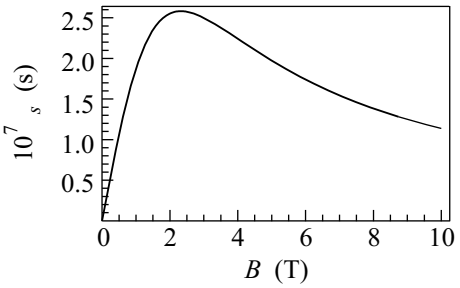
$$a\xi_0 \to a\xi_0'\left(H\right) = a\xi_0\left[1 + \frac{1}{240}\left(a\sqrt{\xi_0^3/2\pi}/R_C\right)^2\right] \qquad (26)$$

(here $a\sqrt{\xi_0^3/2\pi}$ is the size in space of the empty area at the saddle point). As a result,

$$1/\tau_S\left(H\right) = \left(1/\tau_S\right)\exp\left[\frac{1}{240}\left(\frac{a}{R_C}\right)^2\left(\xi_0^3/2\pi\right)^{3/2}\right]. \qquad (27)$$

For GaAs, the exponent in Eq. (27) can be estimated as $0.15\left(B/1\mathrm{T}\right)^2\left(1\mathrm{K}/T\right)^{3/2}\left(10^{11}\mathrm{cm}^{-2}/n_d\right)^{3/4}$.

On Figure, the dependence of the spin relaxation time on the external magnetic field is shown for the combination of the two mechanisms described by Eqs. (15) and (21) and the same parameters that were used for numerical estimates. The maximum is found at $B \approx 2$ T. This dependence is in qualitative agreement with the experimental data obtained in Ref. [9] for small and intermediate magnetic fields.



The spin relaxation time according to mechanisms described by Eqs. (15) and (21)

To conclude, the theory of spin relaxation in the impurity band of a 2D semiconductor in the external magnetic field is presented. It is shown that spin precession in the external magnetic field enhances spin-orbit-induced and suppresses hyperfine-interaction-induced spin relaxation. For spin orbit coupling, the relaxation rate is linear in B over a wide range of parameters. For hyperfine interaction, the spin relaxation rate is inversely proportional to the external magnetic field.

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