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Anisotropic spin diffusion in a semiconductor quantum well

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We show that spin diffusion of an inhomogeneous spin density distribution in an asymmetric zinc-blende semiconductor quantum well is anisotropic in coordinate space, if the D'yakonov-Perel' spin relaxation mechanism is dominant. This anisotropy depends on the relation between the Dresselhaus and Rashba contributions to the spin splitting and reaches its maximum when the both contributions are equal in magnitude. Under this condition the temporal behavior of the spin density strongly depends on the relation between the initial spatial extent of the spin packet and spin diffusion length.

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The temporal and spatial evolution of an inhomogeneous spin polarization in semiconductor quantum well (QW) and heterostructures has attracted much attention in the recent years in connection with the possible use of the spin degrees of freedom in spintronics [1]. It is known that in A_3B_5 semiconductors already for the homogeneous spin polarization there is correlations between the motion of electrons in the coordinate and spin spaces [2 – 4]. These correlations are a consequence of a linear (in 2D structures) dependence of the spin-orbit interaction (SOI) on the electron momentum $\mathbf{k}$. The SOI is also an essential ingredient of the D'yakonov-Perel' (DP) spin relaxation mechanism [5], which is dominant in most A_3B_5 bulk semiconductors and heterostructures.

It is well known, that in a zinc-blende semiconductor QW there are two contributions to the SOI. The first of them arises due to the bulk inversion asymmetry of the material (V_{BIA}) and induces the Dresselhaus spin splitting [6, 7], while the second is caused by the structure inversion asymmetry of the QW itself (V_{SIA}) and leads to the Rashba spin splitting [8]. V_{BIA} is fixed by the choice of material of which the QW is made, but V_{SIA} can be modified by the application of a gate voltage.

In this paper we point to a new phenomena which is due to the interference of V_{BIA} and V_{SIA}. First, we show that in the case when the DP spin relaxation mechanism is dominant the interference of V_{BIA} and V_{SIA} leads to an anisotropy (in the coordinate space) of the *inhomogeneous* spin distribution in the QW plane. Second, we show that the simultaneous presence of V_{BIA} and V_{SIA} leads to a non-exponential relaxation of the spin density. To this end we solve the diffusion equation for the spin density $\mathbf{M}(\mathbf{r}, t)$ assuming that the initial spin distribution is created by a short laser pulse, focused to a spot with a diameter a much less than the spin diffusion length L_s. This condition can be satisfied in case of n-doped semiconductor in which the electrons spin relaxation times are relatively long and the spin diffusion rates are relatively high [9, 10].

We want to emphasize that in case of homogeneous spin polarization and in the absence of the magnetic field V_{BIA} and V_{SIA} give additive contributions to the DP relaxation rate $1/\tau_s$ of the spin polarization initially oriented perpendicularly to the QW [3], that is, $1/\tau_s$ is not sensitive to the relation between V_{BIA} and V_{SIA}. In the external magnetic field $\mathbf{B}$ the contributions of V_{BIA} and V_{SIA} to the spin relaxation become non-additive [11, 12] but the relaxation still remains exponential.

Note that the spin diffusion in a semiconductor QW provided that the DP spin relaxation mechanism is dominant have been considered theoretically in Ref. [13]. However, only V_{SIA} contribution to the SOI was taken into account in the calculation of the spin distribution, which is inevitably isotropic in the coordinate space in that case, if $\mathbf{B} = 0$. The goal of our consideration is different. We want to demonstrate that the interplay between V_{BIA} and V_{SIA} leads to a dependence of the spin density $\mathbf{M}(\mathbf{r}, t)$ on the direction of $\mathbf{r}$ even in the absence of the external magnetic field.

The spin-orbit part of the electron Hamiltonian V_{so} has the form

$$V_{so} = V_{BIA} + V_{SIA} = \frac{\hbar}{2}\Omega(\mathbf{k}) \cdot \sigma, \qquad (1)$$

where $\mathbf{k}$ is the electron wave vector in the QW plane, $\sigma = \{\sigma_x, \sigma_y, \sigma_z\}$ are the Pauli matrices, and $\Omega(\mathbf{k})$ can be regarded as a $\mathbf{k}$-dependent effective magnetic field. In the following we will consider (001) grown QW. In this case the anisotropy of the spin diffusion is more pronounced and, at the same time, allows the most simple theoretical consideration. For a sufficiently narrow QW

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one can neglect cubic in $\mathbf{k}$ terms in $\Omega(\mathbf{k})$ [7]. The vector $\Omega(\mathbf{k})$ lies in the QW plane and its components are given by

$$\Omega(\mathbf{k}) = [(\alpha_1 k_x + \alpha_2 k_y), -(\alpha_1 k_y + \alpha_2 k_x)], \qquad (2)$$

where the coefficients α_1 and α_2 correspond to V_{BIA} and V_{SIA}, respectively.

The diffusion equation for the spin density $\mathbf{M}(\mathbf{r}, t)$ has been presented in Refs. [13, 14]. Here we only discuss the applicability of the diffusion approximation to our problem. The diffusion approximation is applicable if $\tau v_f/a$, $\tau\Omega \ll 1$, where τ is the mean scattering time, a is the size of the spin packet, and v_f is the Fermi velocity. These inequalities mean that the motions in the coordinate and spin spaces, consequently, are diffusive. To give a more quantitative insight into the applicability of the diffusive approximation for a GaAs-based QW, we take a typical value of the Fermi vawevector $k_f = 10^{-6}$ cm^{-1}, which corresponds to the sheet electron density $N = k_f^2/2\pi \approx 2 \cdot 10^{11}$ cm^{-2} and the Fermi velocity $v_f = \hbar k_f/m \approx 10^6$ cm/s, where $m = 0.067 m_e$ is the electron effective mass. As was recently clarified in Ref. [15], when considering the DP spin relaxation mechanism, the mean scattering time τ must be calculated taking into account electron-electron collisions. For this reason, in the case of n-doped semiconductor, τ can be approximately estimated as an optical dephasing time, measured in time-resolved four wave-mixing experiments [16] in the spectral range of the interband transitions. This time is of the order of 0.1 ps and there is no essential difference between the times in 2D and 3D systems [17]. If we take the lower bound for the spot size $a = 1$ μm (the spot size can not be made smaller than the laser wavelength) and the upper bound for the spin splitting $\hbar\Omega = 1$ meV [18], we obtain $\tau v_f/a \sim 10^{-3}$ and $\tau\Omega \sim 10^{-1}$. Thus, the diffusion approximation can be applied to our problem without any essential restrictions.

In n-doped semiconductor the excess of photoexcited electrons and holes rapidly disappears due to the recombination of holes with background electrons. In other words, only the spin packet but not the charge packet is present in the semiconductor [19]. This allows one to neglect the coupling between charge and spin degrees of freedom. Then, for the degenerate electron gas the diffusion equation takes the form [13]

$$\frac{\partial \mathbf{M}(\mathbf{r}, t)}{\partial t} - [D\nabla_{\mathbf{r}}^2 + \hat{\Gamma}]\mathbf{M}(\mathbf{r}, \mathbf{t}) - \\ - [\mathbf{b}(\nabla_{\mathbf{r}}) + \Omega_L] \times \mathbf{M}(\mathbf{r}, t) = \mathbf{I}(\mathbf{r}, t), \qquad (3)$$

where $D = \tau v_f^2/2$ is the diffusion constant, $\Omega_L = g\mu_B \mathbf{B}/\hbar$ is the electron spin resonance frequency in a magnetic field $\mathbf{B}$, $\hat{\Gamma}$ is the tensor of the DP spin relaxation rates, and $\mathbf{I}(\mathbf{r}, t) = [0, 0, I_z(\mathbf{r}, t)]$ is a source of spin-polarized electrons. The operator $\mathbf{b} = \tau k_f v_f \Omega(\nabla_{\mathbf{r}})$ mixes different components of the *inhomogeneous* spin polarization $\mathbf{M}(\mathbf{r}, t)$. In accordance with the typical experimental conditions we assume that the magnetic field is parallel to the QW plane and $\tau\Omega_L \ll 1$.

Following Ref. [13], we perform the Fourier transform of the spin density $\mathbf{M}(\mathbf{r}, t)$ in Eq. (3). The algebraic equation for $\mathbf{M}(\mathbf{q}, \omega)$ takes the form

$$(-i\omega + Dq^2 + \hat{\Gamma})\mathbf{M} + [i\mathbf{b}(\mathbf{q}) + \Omega_L)] \times \mathbf{M} = \mathbf{I}(\mathbf{q}, \omega). \qquad (4)$$

Since rather short ($\sim$ 100 fs) laser pulses are used now in experimental investigations of the spin dynamics it is justified to take the delta-function-type approximation for the time dependence of the source term $\mathbf{I}(\mathbf{r}, t)$ in Eq. (3). However, the same is not always true about the spatial dependence of the source term, since the diameter a of the laser spot and the spin diffusion length L_s may be comparable in magnitude. Moreover, it is interesting to trace the evolution of the main features of the spin diffusion when the ratio a/L_s varies from $\ll 1$ to $\gg 1$. For this reason we take the Gaussian distribution for the initial spin density:

$$I_z(\mathbf{r}, t) = \frac{M_0}{\pi a^2} \exp(-\frac{r^2}{a^2})\delta(t), \qquad (5)$$

$$I_z(\mathbf{q}, \omega) = M_0 \exp(-\frac{a^2 q^2}{4}). \qquad (6)$$

Before solving Eqs. (3) and (4) we discuss their symmetry properties. First of all we note that these equations are not invariant under the spatial inversion, that is, $\mathbf{M}(\mathbf{r}, \mathbf{B}, t) \neq \mathbf{M}(-\mathbf{r}, \mathbf{B}, t)$. However, since the laser source term possesses the symmetry $I_z(\mathbf{r}, t) = I_z(-\mathbf{r}, t)$, the solution of Eq. (3) obeys the exact relation

$$M_z(\mathbf{r}, \mathbf{B}, t) = M_z(-\mathbf{r}, -\mathbf{B}, t). \qquad (7)$$

This relation is a special feature of an inhomogeneous spin distribution which varies with time under the DP spin relaxation mechanism.

Next we note that the tensor $\hat{\Gamma}$ and the vector Ω_L do not depend on $\mathbf{q}$ and unable to create the anisotropy in the coordinate space if $\mathbf{b}(\mathbf{q}) = 0$. Hence, the only source of the anisotropy is the vector $\mathbf{b}(\mathbf{q})$ which enter Eq. (4) through the quantity $\mathbf{h} = i\mathbf{b}(\mathbf{q}) + \Omega_L$. In turn, the vector $\mathbf{h}$ forms two independent invariants, namely, $\mathbf{h}^2$ and $h_i h_j \Gamma_{ij}$, which depend on the direction of $\mathbf{q}$ and determine the anisotropy of $M_z(\mathbf{r}, t)$. In the considered case of the DP spin relaxation mechanism, these invariants have the common microscopic origin. If only one

contribution to the SOI is nonzero ($\alpha_1\alpha_2 = 0$), these invariants are equivalent, since the tensor $\hat{\Gamma}$ is isotropic in the QW plane [11]. In this case the anisotropy of the spin diffusion is entirely due to the external magnetic field, since $|\mathbf{b}(\mathbf{q})|$ does not depend on the direction of $\mathbf{q}$. This situation has been analyzed in Ref. [13].

Here we want to analyze the intrinsic anisotropy of the spin diffusion which is due to the simultaneous presence of V_{BIA} and V_{SIA} and exists in the absence of the external magnetic field. To make the following consideration more transparent we transform the in-plane coordinate system xy to a new coordinate system $x'y'$ where the x' and y' axes are oriented along [110] and [1$\bar{1}$0] directions, respectively. From Eq. (2) we find the components of the vector $\Omega(\mathbf{k})$ in the new coordinate system:

$$\Omega_{x'} = (\alpha_2 - \alpha_1)k_{y'}, \tag{8}$$

$$\Omega_{y'} = -(\alpha_2 + \alpha_1)k_{x'}. \tag{9}$$

If V_{BIA} or V_{SIA} equals zero ($\alpha_1\alpha_2 = 0$), the spin splitting is isotropic and, as a consequence, the diffusion is isotropic too. This case has been analyzed in [13]. In the opposite case of strong anisotropy ($\alpha_1 = \pm\alpha_2$) the vector $\Omega(\mathbf{k})$ is oriented along the [1$\bar{1}$0] or [110] directions, respectively.

Only numerical solution of Eq. (3) is possible in case of arbitrary relation between α_1 and α_2. Since our prime interest is in the anisotropy of the spin distribution, we consider the ultimate case $\alpha_1 = \alpha_2$ when $\Omega(\mathbf{k}) \parallel [1\bar{1}0]$ and the anisotropy becomes as large as possible. Fortunately, in this case Eq. (3) has a simple analytical solution.

To solve Eqs. (3) and (4) we also need to know the components of the tensor $\hat{\Gamma}$. As shown in Ref. [11], when both V_{BIA} and V_{SIA} are nonzero, the tensor $\hat{\Gamma}$ has one normal to the QW plane eigenvector with corresponding relaxation rate $1/\tau_s = \tau k_f^2(\alpha_2^2 + \alpha_1^2)$ and two in-plane eigenvectors oriented along the directions [110] and [1$\bar{1}$0], with the relaxation rates $1/\tau_\pm = \tau k_f^2(\alpha_1 \pm \alpha_2)^2/2$, respectively. Consequently, the anisotropy of the spin relaxation in the QW plane depends on the relation between V_{BIA} and V_{SIA}. Note that the spin relaxation anisotropy manifests itself only if the spin density $\mathbf{M}$ is not orthogonal to the QW plane. Hence, it is not obvious that this in-plane anisotropy of the spin relaxation must necessarily lead to the anisotropy of the spin diffusion if the initially created spin density $\mathbf{M}(\mathbf{r}, t = 0)$ is orthogonal to the QW plane.

Since we are interested in the intrinsic anisotropy of the spin distribution, rather than one due to the external magnetic field, we consider only the special case when the magnetic field $\mathbf{B} \parallel [1\bar{1}0]$, that is, directed along $\Omega(\mathbf{k})$. In this case $M_{y'} = 0$ since both $\mathbf{B}$ and $\Omega(\mathbf{k})$ are directed along y' and unable to create $M_{y'}$ if the initial spin polarization is oriented along z. Solving Eq. (4) for $\mathbf{M}_z(\mathbf{q}, \omega)$ and performing the inverse Fourie transform we obtain

$$M_z(\mathbf{r}, t) = \int \frac{d\mathbf{q}}{(2\pi)^2} I_z(\mathbf{q})(e^{-\lambda_1(\mathbf{q})t} + e^{-\lambda_2(\mathbf{q})t})e^{i\mathbf{qr}}, \tag{10}$$

where $\lambda_{1,2} = i\omega_{1,2}$ are the eigenvalues of Eq. (4)

$$\lambda_{1,2}(\mathbf{q}) = Dq^2 + \frac{1}{\tau_s} \pm 2\frac{L_s}{\tau_s}q_{x'} \pm i\Omega_L, \tag{11}$$

where $L_s = \sqrt{D\tau_s}$ is the spin diffusion length. Inserting the expression (6) for $I_z(\mathbf{q})$ into the integrand of Eq. (10) and performing the integration we get

$$M_z(\mathbf{r}, t) = \frac{M_0}{F}\exp\left(-\frac{r^2}{F} - \frac{t}{\tau_s}\frac{a^2}{F}\right)\cos\left(4\frac{L_s}{\tau_s}\frac{t}{F}x' - \Omega_L t\right), \tag{12}$$

where $F = 4Dt + a^2$. From this equation we notice that the spin density distribution $M_z(\mathbf{r}, t)$ is anisotropic. The isotropic exponential decrease of $M_z(\mathbf{r}, t)$ with r^2 is accompanied by the spatial oscillations of the spin density along the x' axis.

A distinctive feature of Eq. (12) is the very simple dependence of $M_z(\mathbf{r}, t)$ on the magnetic field. The spin density oscillates (in addition to the diffusive relaxation) with the frequency Ω_L at any point $\mathbf{r}$ and there is no threshold value $\Omega_L \sim 1/\tau_s$ (see e.g. Ref. [13]) for the frequency of these oscillations. Of course, this peculiarity is a consequence of the special choice $\mathbf{B} \parallel \Omega(\mathbf{k}) \parallel [1\bar{1}0]$ we take in our consideration. If these restrictions will relax, the evolution of the spin density will become much more complicated. We will not go into details of the influence of the magnetic field on the anisotropy of the spin diffusion. It seems likely that the transverse magnetic field is not especially useful when studying the anisotropy of the inhomogeneous spin distribution, since this field itself is a source of the anisotropy. This topic requires an additional analysis and is outside the scope of this paper.

The general case, Eq. (12), of arbitrary relation between the spot size a and the spin diffusion length L_s is rather complicated. To give a more physical insight into the spin diffusion, let us consider two limiting cases of the small and large laser spot size a as compared with the spin diffusion length L_s. If $a \gg L_s$, then $F \approx a^2$ and Eq. (12) gives the well known [9] decaying oscillations of the homogeneous spin density

$$M_z(t) \approx \frac{M_0}{a^2}\exp\left(-\frac{t}{\tau_s}\right)\cos(\Omega_L t). \tag{13}$$

In order to understand better the opposite limit of strong inhomogeneity $a \ll L_s$ we formally set $a = 0$ in Eq. (12). Then $F = 4Dt$ and we get from Eq. (12)

$$M_z(\mathbf{r}, t) \approx \frac{M_0}{4Dt} \exp\left(-\frac{r^2}{4Dt}\right) \cos\left(\frac{x'}{L_s} - \Omega_L t\right). \qquad (14)$$

The temporal behavior of the spin density, given by this equation, is distinctly different from that of the isotropic spin distribution [13] (in zero magnetic field) since the combination t/τ_s does not enter Eq. (14) at all. Instead, *at every point* $\mathbf{r}$ much more slow diffusive relaxation occurs. In particular, at the center of the laser spot $M_z \propto 1/t$. To obtain the physically meaningful spin density $M_z(\mathbf{r}, t)$ we must substitute in Eq. (12) small but finite $a \approx \lambda$, where λ is the laser wavelength. A more careful analysis of Eq. (12) shows that the exponential decrease with time of the spin density at $\mathbf{r} = 0$ still persists at small times $t \lesssim \tau_s(a/L_s) \ll \tau_s$ and only then the spin density decreases as $1/t$. Thus, Eq. (14) is valid at all but small times $t \ll \tau_s$.

The spin relaxation time τ_s enters Eq. (14) only through the spin diffusion length L_s, which determines in this case the scale of the anisotropy variation. It is seen from this equation that the pronounced anisotropy of the spin distribution appears at the distances $r \gtrsim L_s$ from the laser spot.

The evolution of the inhomogeneous spin density is a result of the spin precession in the angular space and the spin diffusion in the coordinate space. When the anisotropy of the spin splitting is strong the interplay between the gradient of the spin density and the spin precession (see the term with the operator $\mathbf{b}$ in Eq. (3)) leads to the unusual temporal behavior of the spin density given by Eq. (14). It is instructive to see the temporal evolution of the *total* spin polarization of the QW. Integrating Eq. (14) over the QW plane we obtain immediately

$$\int M_z(\mathbf{r}, t) d\mathbf{r} \sim M_0 \exp\left(-\frac{t}{\tau_s}\right) \cos\ (\Omega_L t). \qquad (15)$$

Thus, the total spin polarization undergoes the exponentially decaying oscillations as in the homogeneous case irrespective of the relationship between a and L_s. This clearly demonstrates that the unusual temporal behavior of Eq. (14) is a consequence of the spatial redistribution of the spin density, which, in turn, arises due to the anisotropy of the spin diffusion.

In conclusion, we have shown that in the (001) grown zinc-blende semiconductor QW the anisotropy of the spin splitting, resulting from the simultaneous presence of the bulk and structure inversion asymmetries, leads to the anisotropic in the QW plane spin diffusion. This anisotropy manifests itself most strongly if the Dresselhaus and Rashba spin splittings are comparable in magnitude and if the initial size of the spin packet is much less as compared to the spin diffusion length. Though our calculations refer to the special case of the equal Dresselhaus and Rashba spin splittings they point to the trend of how the spin distribution changes when the anisotropy of the spin splitting appears. To observe the predicted behavior of the spin density given by Eq. (14) it is advantageous to use experimental technique similar to that in Ref. [20], where the time-resolved Faraday rotation in an n-doped GaAs/AlGaAs QW was measured while applying a bias to the gate electrode. Among other things, the gate voltage can be used to modify the Rashba spin splitting thereby creating the interference between V_{BIA} and V_{SIA}. We believe that experimental observations of the anisotropic spin diffusion can give an additional information on the relation between V_{BIA} and V_{SIA}. Besides, we have shown, that the interference affects not only the anisotropy of the spin density distribution but also the temporal evolution of the spin density at $\mathbf{r} = 0$, measured at the overlapping pump and probe laser spots. Hence, the decay of the spin density with time (in time resolved experiments) can deviate from a purely exponential when the spin relaxation rate is slow and, as a consequence, the spin diffusion length is large. This should be kept in mind when extracting the spin relaxation times from experimental data. We would like to draw attention to the symmetry relation given by Eq. (7), which is also accessible to experimental verification. This relation is not connected with the interference of V_{BIA} and V_{SIA}, but is an inherent characteristic of the DP spin relaxation mechanism. Hence, it can help in the discrimination between different spin relaxation mechanisms. As the symmetry relation Eq. (7) is characteristic for DP spin relaxation, the violation of this relation would imply that DP spin relaxation is not dominant.

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