Spin oscillations in transient diffusion of a spin pulse in *n*-type semiconductor quantum wells

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By studying the time and spatial evolution of a spin-polarized pulse in *n*-type semiconductor quantum wells, we highlight the importance of the off-diagonal spin coherence in spin diffusion and transport. Spin oscillations and spin polarization reverse along the direction of spin diffusion in the absence of the applied magnetic field are predicted from our investigation.

There are growing numbers of experimental and theoretical investigations on spin related phenomena due to potential applications in semiconductor spintronic devices such as spin filters and spin transistors.[1–7] An important prerequisite for the realization of such devices is to understand how the spin-polarized electrons transport from one position to another. In experiment, coherent spin propagation over a long distance has been reported.[8] Spin injections from ferromagnetic materials into semiconductors have been extensively investigated.[9] In theory most works are based on the quasi-independent electron model and are focused on the diffusive transport regime[2,10] in which the spin polarization of the current is controlled by the longitudinal spin dephasing through a relaxation-time approximation. There are some efforts on investigating the spin dephasing in the spin transport: Takahashi *et al.* calculated the spin-diffusion coefficients by solving the kinetic equations with only the electron-electron scattering.[11] Bournel *et al.*[12] and Saikin *et al.*[13] studied the spin transport in semiconductor heterostructures with the Rashba effect and the electron-phonon scattering by Monte Carlo simulation. However, in these investigations either the off-diagonal inter-spin-band correlations $\rho_{\mathbf{k}\sigma-\sigma} = \langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}-\sigma} \rangle$ are discarded (or not explicitly included) or the spin dephasing is simply introduced through the relaxation-time approximation.

Recently we performed a many-body investigation of spin transport in GaAs (100) quantum wells (QW's) by self-consistently solving the many-body kinetic transport equations together with the Poission equation. We have explicitly taken into account the spatial inhomogeneity, the D'yakonov-Perel' (DP) mechanism, and all scattering.[14] In our theory, both the diagonal intra-spin-band correlations, i.e., the electron distribution function $f_{\mathbf{k}\sigma} = \langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} \rangle$, and the off-diagonal inter-spin-band correlations $\rho_{\mathbf{k}\sigma-\sigma}$ are explicitly included. The spin dephasing time, the spin/charge diffusion length together with the mobility are calculated self-consistently. We further pointed out a spin dephasing mechanism in the spin diffusion/transport that the inhomogeneous broadening due to the interference between the electrons/spins with different momenta along the direction of the diffusion can cause spin dephasing in the presence of the scattering and the resulting dephasing can be more important than the dephasing due to the DP mechanism. In this paper, we further investigate the time evolution of a spin-polarized pulse (SPP) in *n*-type GaAs (100) QW through the many-body kinetic Bloch equations. We highlight the importance of the *off-diagonal spin correlations* in the spin diffusion and transport.

In *n*-type GaAs QW's, the dominant spin dephasing mechanism is the DP mechanism.[15,16] By taking account of the DP term, the kinetic Bloch equations can be written as

$$\frac{\partial \rho(\mathbf{R},\mathbf{k},t)}{\partial t} - \frac{1}{2}\{\boldsymbol{\nabla}_{\mathbf{R}}\bar{\varepsilon}(\mathbf{R},\mathbf{k},t), \boldsymbol{\nabla}_{\mathbf{k}}\rho(\mathbf{R},\mathbf{k},t)\}$$

$$+ \frac{1}{2}\{\boldsymbol{\nabla}_{\mathbf{k}}\bar{\varepsilon}(\mathbf{R},\mathbf{k},t), \boldsymbol{\nabla}_{\mathbf{R}}\rho(\mathbf{R},\mathbf{k},t)\} - \frac{\partial \rho(\mathbf{R},\mathbf{k},t)}{\partial t}\bigg|_{c}$$

$$= \frac{\partial \rho(\mathbf{R},\mathbf{k},t)}{\partial t}\bigg|_{s}. \tag{1}$$

Here $\rho(\mathbf{R},\mathbf{k},t)$ represents a single-particle density matrix. The diagonal elements describe the electron distribution functions $\rho_{\sigma\sigma}(\mathbf{R},\mathbf{k},t) = f_{\sigma}(\mathbf{R},\mathbf{k},t)$ of wave vector $\mathbf{k}$ and spin $\sigma(=\pm 1/2)$ at position $\mathbf{R}$ and time t. The off-diagonal elements $\rho_{\sigma-\sigma}(\mathbf{R},\mathbf{k},t)$ describe the inter-spin-band correlations (coherences) for the spin coherence. The quasiparticle energy $\bar{\varepsilon}_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t)$, in the presence of a moderate magnetic field $\mathbf{B}$ and with the DP term[15] included, reads $\bar{\varepsilon}_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t) = \varepsilon_k \delta_{\sigma\sigma'} + [g\mu_B\mathbf{B} + \mathbf{h}(\mathbf{k})] \cdot \boldsymbol{\sigma}_{\sigma\sigma'}/2 - e\psi(\mathbf{R},t) + \Sigma_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t)$, where $\varepsilon_k = k^2/2m^*$ is the energy spectrum with m^* denoting the electron effective mass. $\boldsymbol{\sigma}$ are the Pauli matrices. $\mathbf{h}(\mathbf{k})$ denotes the effective magnetic field from the DP effect which contains contributions from both the Dresselhaus[17] term and the Rashba term.[18] In this paper, we only consider the Dresselhaus term which can be written as[19] $h_x(\mathbf{k}) = \gamma k_x(k_y^2 - \kappa_z^2)$ and $h_y(\mathbf{k}) = \gamma k_y(\kappa_z^2 - k_x^2)$ with γ denoting the spin-orbit coupling strength[20] and κ_z^2 representing the average of the operator $-(\partial/\partial z)^2$ over the electronic state of the lowest subband. The electric potential $\psi(\mathbf{R},t)$ satisfies the Poission equation

$$\boldsymbol{\nabla}_{\mathbf{R}}^2 \psi(\mathbf{R},t) = -e[n(\mathbf{R},t) - n_0(\mathbf{R})]/\epsilon, \tag{2}$$

where $n(\mathbf{R},t) = \Sigma_{\sigma\mathbf{k}} f_{\sigma}(\mathbf{R},\mathbf{k},t)$ is the electron density at position $\mathbf{R}$ and time t and $n_0(\mathbf{R})$ is the positive background electric charge density. $\Sigma_{\sigma\sigma'}(\mathbf{R},\mathbf{k},t) = -\Sigma_{\mathbf{q}} V_{\mathbf{q}} \rho_{\sigma\sigma'}(\mathbf{R},\mathbf{k}-\mathbf{q},t)$ is the Hartree-Fock self-energy, with $V_{\mathbf{q}}$ standing for the Coulomb matrix element. In two-dimensional case, $V_{\mathbf{q}}$ is given by $V_{\mathbf{q}} = 2\pi e^2/[\epsilon_0(q+\kappa)]$, with $\kappa = (2e^2m^*/\epsilon_0)\Sigma_{\sigma} f_{\sigma}(\mathbf{k}$

$=0$) being the inverse screening length. ϵ_0 represents the static dielectric constant. It is noted that if one only takes account of the diagonal elements $\rho_{\sigma\sigma}$ and neglects all the off-diagonal elements $\rho_{\sigma-\sigma}$ in Eq. (1), the first three terms on the left-hand side of the equation correspond to the driving terms in the classical Boltzmann equation, modified with the DP term and the self-energy term from the Coulomb Hartree contribution. $\partial\rho(\mathbf{R},\mathbf{k},t)/\partial t|_c$ and $\partial\rho(\mathbf{R},\mathbf{k},t)/\partial t|_s$ in the Bloch equations (1) are the coherent and scattering terms, respectively. The coherent term describes the electron spin precession around the applied magnetic field and the effective magnetic field from the DP term. Its expression as well as that of the scattering term $\partial\rho(\mathbf{R},\mathbf{k},t)/\partial t|_s$ are given in detail in Refs. 14 and 21.

It is noted that by using a spin-flip time τ_{sf} to describe the spin dephasing caused by the DP term and summing over the momentum, one is able to derive the diffusion equations for charge and spin densities of electrons in the so-called "mean-field" approximation.[2,22] However, as pointed out in our previous papers,[14] the adoption of the mean-field approximation removes the interference between the electrons with different momentums and thus overlooks the inhomogeneous broadening that causes additional spin dephasing in the spin transport. We further point out in this paper that by using the spin-flip time approximation, some of the most marked features of the DP mechanism are thrown away.

We assume that at initial time $t=0$ there is a SPP centered at $x=0$. The electrons are locally in equilibrium, i.e., $f_\sigma(x,\mathbf{k},0)=(\exp\{[(\epsilon_k-\mu_\sigma(x)]/T_e\}+1)^{-1}$, where $\mu_\sigma(x)$ stands for the chemical potential of electrons with spin σ at position x and is determined by the corresponding electron density: $N_\sigma(x,0)=\Sigma_k f_\sigma(x,\mathbf{k},0)$. The shape of the initial spin pulse is assumed to be Gaussian like

$$\Delta N(x,0)=N_{1/2}(x,0)-N_{-1/2}(x,0)=\Delta N_0 e^{-x^2/\delta x^2}, \quad (3)$$

with ΔN_0 and δx representing the peak and width of the SPP, respectively. We further assume that there is no spin coherence at the initial time, $\rho_{\sigma-\sigma}(x,\mathbf{k},0)=0$. This SPP can be achieved by a circularly polarized laser pulse.

By numerically solving the kinetic equations (1) together with the initial conditions and the Poission equation (2), we are able to study the temporal evolution of the SPP with $T=200$ K and $B\equiv0$. A typical calculation of the spin diffusion in n-type GaAs QW is carried out by choosing the total electron density $N_e[\equiv n_0(\mathbf{R})]=4\times10^{11}$ cm^{-2}, the maximum spin imbalance in Eq. (3) $\Delta N_0=1\times10^{11}$ cm^{-2} and the width of the spin pulse $\delta x=0.15$ μm. The material parameters are taken from Ref. 23. The absolute value of the spin imbalance $|\Delta N(x,t)|$ and the incoherently summed spin coherence[24] $\rho=\Sigma_\mathbf{k}|\rho_{\mathbf{k}\sigma-\sigma}(\mathbf{R},t)|$ are plotted as functions of the position x along the diffusion direction and the time t in Figs. 1 (a) and 1(b), respectively for impurity-free case ($N_i=0$). Here N_i denotes the impurity density. One can see from the figure that due to the strong diffusion as well as the spin dephasing, the spin polarization at the center of the spin pulse decays very fast initially. In the mean time, the spin coherence ρ goes up due to the precession of spins in the presence of the effective magnetic field of the DP term. After
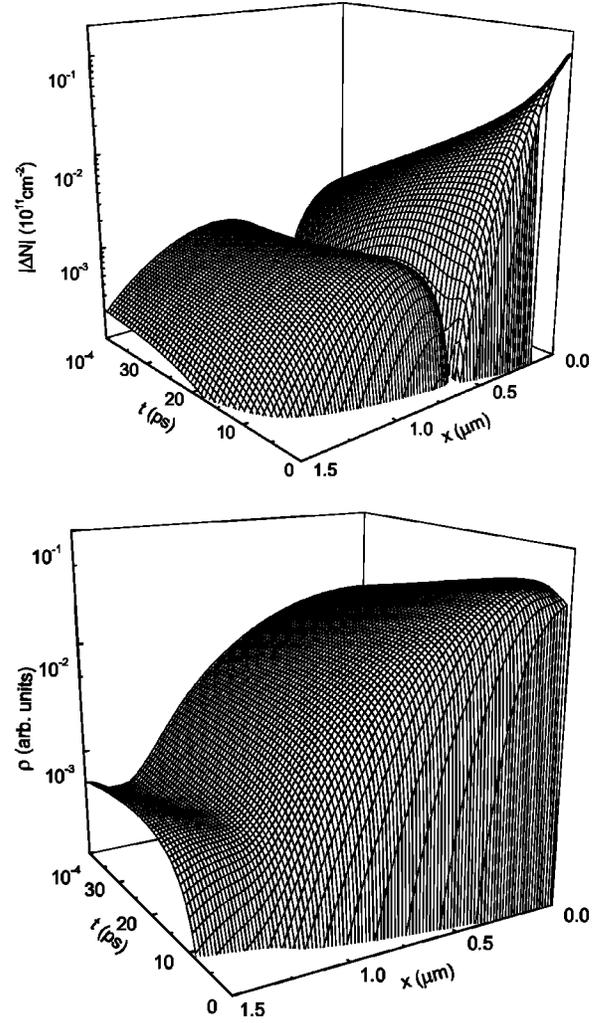

FIG. 1. The absolute value of the spin imbalance $|\Delta N|$ and the incoherently summed spin coherence ρ vs the position x and the time t for the impurity-free ($N_i=0$) case.

10 ps, as the diffusion becomes weaker due to the smaller spatial gradient, the decay rate at the center of the spin signal slows down and the spin coherence begins to decay due to the diffusion as well as the spin dephasing. It is noted that the spin polarization away from the center, e.g., in the region 0.12 μm $<x<0.15$ μm, first increases due to the net spin diffusion from the center and then decays after the diffusion from the center becomes moderate. For the region out of the initial spin pulse ($x>0.15$ μm), the combined effect of the diffusion and the dephasing leads to more complicated behaviors. The most striking feature of the evolution of the SPP is that the spin polarization can be *opposite* to the initial one even in the *absence* of the applied magnetic field and there are oscillations in the time evolution of the spin polarization at some positions.

From Fig. 1(a) one can see that there is another peak in the spin polarization at the positions out of the initial spin pulse after 10 ps. However the spin polarization of this second peak is *opposite* to the initial one. As the time goes on, the second peak becomes larger due to the decay of the first peak in the center. After 30 ps, the heights of these two peaks
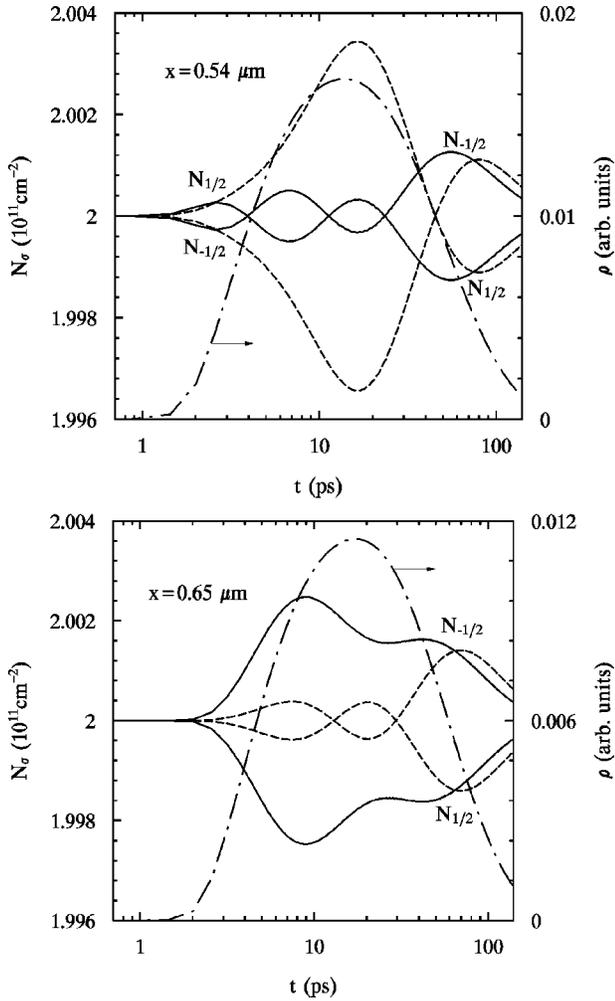
FIG. 2. The electron densities for different spins at (a) x $=0.54$ μm and (b) $x=0.65$ μm vs time t with solid curves for N_i $=0$ and dashed ones for $N_i=0.1N_e$. The dotted curves are the corresponding incoherently summed spin coherence at the same position for $N_i=0$ case.

are comparable. Moreover, in the region 0.5 μm$<x$ <0.7 μm, there are oscillations in the time evolution of spin polarization. The details are shown in Fig. 2 where the densities of the electrons with different spin $N_\sigma(x,t)$ are plotted at two typical positions $x=0.54$ and 0.65 μm for the cases with (dashed curves) and without (solid curves) impurities.

It is seen from Fig. 2(a) that at $x=0.54$ μm for impurity-free case, there is no spin signal in the first picosecond as the position is located out of the initial spin pulse. Then the spin signal starts to build up due to the diffusion of both the diagonal electron distribution $f_\sigma(x,\mathbf{k},t)$ and the off-diagonal spin coherence $\rho_{\mathbf{k}\sigma-\sigma}(x,t)$ from the center. With the joint effects of the DP term as well as the diffusion, the spin signal reaches its first peak at about 2.5 ps and then decreases. It gets a crossing of spin-up and -down electron densities at 4 ps. After that the spin-down electrons exceed the spin-up ones and hence the spin polarization changes sign. The difference between the spin-up and -down electrons further increases until 7 ps when the difference arrives at another peak. The oscillations go on with the periods becoming

larger and larger. After the third oscillation, the period becomes too long to observe any oscillation in the time regime of our calculation. As a result after the last crossing at 25 ps, the spin density of spin-down electrons is larger than that of the spin-up ones and the spin polarization is reversed. One can further see from the figure that, in the regime when the polarization oscillates, the spin coherence keeps increasing. This indicates that the spin coherence ρ at this position mainly comes from the diffusion from the center. It is seen from the coherent terms of the Bloch equations that the imbalance of the distribution functions with the opposite spin and the spin coherence can transfer to each other by the DP effective magnetic field. Therefore, the additional spin coherence ρ diffused from the center induces the spin-polarization oscillations at position outside the initial spin pulse.

It is further noted that the oscillations differ in positions. For example, as shown in Fig. 2(b), at $x=0.65$ μm (and positions farther away), the oscillations are almost marginal but the feature that the spin polarization after a long time is opposite to the initial one remains. Moreover it is noted that in Fig. 2(a), when the spin pulse arrives, its polarization points to the initial one. Nevertheless, in Fig. 2(b) for positions farther away from the initial pulse, it is interesting to see that the spin signal arrives with spin polarization opposite to the initial one. This is understood that at positions far away from the initial pulse, what diffused from the center of the spin pulse is the off-diagonal spin coherence and the diagonal spin imbalance is quite small already at the edge of the initial pulse and therefore its diffusion to the outer space is marginal. The off-diagonal spin coherence induces the reverse of the spin polarization as what is said above. This can be seen from the fact that for $t<10$ ps in Fig. 2(b), both diagonal and off-diagonal terms increase with time. For positions farther away from the center, the arrival of the spin polarization also includes the diagonal components but with the opposite spin polarization.

We now explore the effect of the impurity to the spin diffusion. In Fig. 2 we also plot the corresponding curves of electron densities with impurity density $N_i=0.1N_e$ as dashed curves. The figure shows that the impurities dramatically change the behavior of the spin diffusion but do not eliminate the spin oscillations and the reverse of the spin polarization when time is long enough. By comparing the solid curves with the dashed ones it is noted that when time is long enough, say around 100 ps, the spin polarization for the case with impurities is always larger than that without impurities. This is understood that for the impurity-free case, the mobility of electrons is larger and spin polarization is easier to diffuse away from the center. Moreover, the nonmagnetic impurities tend to retain the spin coherence. This is because the impurity scattering drives the electrons to a homogeneous state in the momentum space and therefore counters the effect of the DP term that drives the electrons to an inhomogeneous state and leads to spin dephasing.[16,21,24]

A similar spin oscillation without an applied magnetic field has been reported by Brand et al.[25] Nevertheless it is noted that it is quite different from what is discussed here. In the work of Brand et al., the spin oscillation happens in a spacial homogeneous system, and the oscillation is due to the

breakdown of the assumption of the collision domination in the spin dephasing at extreme low temperature (a few kelvin). For temperature higher than 10 K, the oscillation disappears as the electron collision rate increases and the assumption of the collision domination recovers. In our work, the spin oscillation comes from the diffusion (spacial gradient) and happens *outside* the initial spin pulse at very high temperature (200 K). The origin of the spin oscillation is therefore by no means only due to the small electron collision rates as in the case of Brand *et al.*, but due to the combination of the diffusion and the precession of spin signals: At positions just outside of the initial spin pulse, the off-diagonal spin coherences of electrons with large momentums first reach there. As the DP term is proportional to the momentum, this fraction of electrons also share large precession frequencies. Therefore it is possible for the spin signal to oscillate at first few picoseconds and in the region just outside spin pulse when the collisions do not affect the momentum of electrons dramatically. As time passes by, the electrons with relatively smaller momentums, consequently with smaller precession frequencies, also arrive at the region where the oscillation occurs. Therefore due to the joint effects of the diffusion as well as the increasing impact of the collisions, the oscillation becomes slower and slower and totally vanishes after a few hundred picoseconds. For the spin signals in the center of the initial pulse there is no spin oscillation for the temperature of our investigation. This coincides with the results of Brand *et. al.*[25] as well as what we discovered in high-temperature cases.[21]

The reverse and oscillation of the spin polarization of a spin pulse along the diffusion in the absence of the applied magnetic field can only be achieved when *both* the diagonal and off-diagonal terms of electron density matrix and the precession caused by the DP term are considered. Once the relaxation-time approximation is adopted to describe the effect of the DP term or the off-diagonal term is dropped, the spin signal at the positions outside the initial SPP first increases due to the diffusion from the spin pulse then decreases monotonically due to the diffusion as well as the dephasing and the spin polarization never changes the sign. This is because in the framework of the relaxation-time approximation, the most important difference between the spin dephasing caused by the DP mechanism and that caused by the other mechanisms is wiped off. Other spin dephasing mechanisms such as the Elliot-Yafet mechanism[26] and the Bir-Aronov-Pikus mechanism[27] cause the spin dephasing through instantaneous spin-flip scattering. However, the DP term acts as a momentum-dependent magnetic field around which the electron spins precess. This precession results in an inhomogeneous broadening in spin phases and leads to spin dephasing in the presence of the scattering. In addition to the dephasing, it is also possible that the precession may switch the magnetic momentum in the transport even without the applied magnetic field and in the high-temperature regime. For some special positions, the combined effect of the diffusion and the precession may lead the spin to oscillate as shown in Fig. 2.

In conclusion, we perform a study of transient spin diffusion of a SPP in *n*-type GaAs QW's by self-consistently solving the full kinetic Bloch equations in which both the diagonal terms, the distribution functions, and the off-diagonal terms, the inter-spin-band spin coherences, of the density matrix are included. We show that by taking the effect of the DP term to the diagonal and off-diagonal terms of the density matrix into account, the spin polarization outside the initial SPP can be reversed even without magnetic field. Moreover, for some special positions, the spin signals oscillate with time. The reverse and oscillations of the spin signals in spin diffusion/transport at positions outside the initial spin pulse have not been reported either theoretically or experimentally for *n*-type semiconductors and are understood from the diffusion of the off-diagonal terms of the electron-density matrix which oscillates due to the precession of electron spins around the effective magnetic field due to the DP effect. We also stress that these features can only be achieved when the *off-diagonal terms* of the electron-density matrix are included *explicitly* in the theory.

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