Spin-Orbit-Mediated Spin Relaxation in Graphene

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We investigate how spins relax in intrinsic graphene. The spin-orbit coupling arises from the band structure and is enhanced by ripples. The orbital motion is influenced by scattering centers and ripple-induced gauge fields. Spin relaxation due to Elliot-Yafet and Dyakonov-Perel mechanisms and gauge fields in combination with spin-orbit coupling are discussed. In intrinsic graphene, the Dyakonov-Perel mechanism and spin flip due to gauge fields dominate and the spin-flip relaxation time is inversely proportional to the elastic scattering time. The spin-relaxation anisotropy depends on an intricate competition between these mechanisms. Experimental consequences are discussed.

Graphene can be useful in future advanced applications because of the reduced dimensionality, the long mean free paths and phase coherence lengths, and the control of the number of carriers [1]. Among possible applications, graphene is investigated as a material for spintronic devices [2, 3, 4, 5, 6, 7, 8]. Spintronics aims to inject, detect, and manipulate the electron spin in electronic devices.

Spin manipulation via the spin-orbit (SO) coupling has been extensively discussed in semiconductors and metals [9]. The SO coupling enables electric, and not just magnetic, control of the spin [10]. In two dimensional (2D) semiconducting structures, inversion asymmetry results in the Rashba SO coupling [11]. Additionally, bulk inversion asymmetry in AlB$_3$ compounds causes the Dresselhaus SO coupling [12]. Device performance is limited by spin relaxation and understanding its origin enables enhanced spin control. Two mechanisms of spin relaxation discussed in the literature [9, 13], the Elliot-Yafet [14, 15] and Dyakonov-Perel [16, 17] mechanisms, can be relevant in graphene.

Elliot-Yafet (EY) spin relaxation is related to how the spin changes its direction during a scattering event [14, 15]. This is possible because the SO coupling produces electronic wave functions that are admixtures of spin and orbital angular momentum. Dyakonov-Perel (DP) [16, 17] spin relaxation is related to spin precession between scattering events by the effective (Zeeman) magnetic field induced by the SO coupling. This SO induced effective (Zeeman) magnetic field changes direction during scattering. In the EY mechanism, the spin relaxation time is proportional to the elastic scattering time $\tau_{el}$, $\tau_{SO}^{EY} \propto \tau_{el}$, whereas the dependence is opposite $\tau_{SO}^{DP} \propto (\tau_{el})^{-1}$ for the DP mechanism. This qualitative difference allows detection of these two competing mechanisms in disordered samples.

Recently, spin transport and spin relaxation were studied in relatively dirty graphene samples [3, 18]. A spin relaxation length $\lambda_{sf} \sim 2 \mu m$ was measured at room temperature and it was indicated that $\lambda_{sf}$ is proportional to the elastic mean free path $l_{e}$, suggesting the EY mechanism to be dominant [3, 18]. The measured spin relaxation length is weakly anisotropic, such that spins “out of plane” relax 20% faster than spins “in plane” [18]. These experiments motivate a study to see if known spin-relaxation mechanisms, or possibly novel effects, dominate spin scattering in graphene.

In this Letter, we consider spin relaxation in intrinsic graphene arising from three ingredients: (i) The SO interaction arises from the band structure and can be enhanced by graphene corrugations, (ii) spin isotropic scattering centers cause momentum relaxation, (iii) topological lattice disorder induce gauge fields that change...
the orbital motion. First, we study the Elliot-Yafet and Dyakonov-Perel methods for this model ignoring effects of gauge fields. Second, we find that a unique interplay of SO and gauge fields (GF) due to topological disorder causes spin relaxation. Our main findings are that DP and GF mechanisms are comparable and dominate the EY mechanism in intrinsic graphene. Interestingly, the DP mechanism implies that spins out of plane relax twice as fast as spins in plane, but GF exhibits the opposite behavior, spins in plane relax faster than spins out of plane (see Fig. 1 for details). The spin-relaxation anisotropy depends on the intricate competition between the DP mechanism and GF. Our results are valid for relatively clean graphene samples where e.g. adatoms do not alter the SO coupling.

Spin-orbit coupling.- There are Rashba and Dresselhaus “like” SO interactions in graphene and we disregard the latter [10, 19, 20]. The total Hamiltonian reads (ℏ = 1)

$$\mathcal{H} = \pm v_F k (\hat{n} \cdot \hat{\sigma}) - \frac{\Delta}{2} (\hat{\sigma} \times \hat{s})_z ,$$

where = +(-) corresponds to the K(K’) point, the spinor basis for K is \(\Psi_K = (A_1, A_1, B_1, B_1)^T\) and for K’ the components are reversed, \(\Psi_{K’} = (B_1, B_1, A_1, A_1)^T\), A and B denote the lattice sites and ↑ and ↓ the electron spin. With this choice, the SO coupling is identical for K and K’, \(\hat{n} = (\cos \theta, \sin \theta, 0)\), tan θ = k_y/k_x, k = \(\sqrt{k_x^2 + k_y^2}\) is quasi-particle momentum with respect to the K(K’) corners of the hexagonal Brillouin zone, \(\Delta = \Delta_{\text{curv}} + \Delta_{\text{curv}}\)

is the dominant Rashba SO coupling constant induced by curvature and/or an external electric fields [19] and \(\hat{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y)\), \(\hat{s} = (\hat{s}_x, \hat{s}_y)\) correspond to Pauli matrices in sublattice and spin space, respectively. The eigenstates of (1) for the K valley are

$$|\Psi_{K, i\xi}\rangle = N_{i\xi} \left[ \left( \begin{array}{c} c_{-\xi} e^{i\phi} \\ c_{\xi} e^{i\phi} \\ e^{-i\xi} \\ e^{i\xi} \end{array} \right) |↑\rangle + \left( \begin{array}{c} c_{\xi} e^{i\phi} \\ c_{-\xi} e^{i\phi} \\ e^{i\xi} \\ e^{-i\xi} \end{array} \right) |↓\rangle \right] e^{i\xi x}$$

with energies \(E_{k\xi}\) = \(s\Delta/2 + \xi D, D = (v_F k)^2 + \Delta^2/4\), where s = ± corresponds to the ↑ (↓) spin states and \(\xi = ±\) denotes the pseudospin degeneracy, \(c_{\xi} = \sqrt{E_{k\xi}/(2D)}\), \(c_{-\xi} = \sqrt{E_{k\xi}/(2D)}\), \(N_{i\xi} = \sqrt{2E_{k\xi}/(v_F k)}\). These eigenstates are polarized in plane of the graphene layer. The spin precession length is \(l_{\text{pre}} = 2\pi v_F/\Delta\).

Elliot-Yafet mechanism.- The “Rashba” SO coupling can change the spin orientation during a scattering event as is typical for the EY mechanism. We study this effect by a decomposition into partial waves with a well defined orbital angular momentum momentum in [21]. Neglecting mixing of the K and K’ valleys, an incoming wave with total angular momentum, \(L = \tilde{L} i\delta_0 \pm s_z/2 + s_z/2\), is an eigenstate of Eq. (1):

$$\Psi_{i\xi K}^\pm (r, \theta) \equiv \left( \begin{array}{c} c_{-\xi} J_n(kr) e^{i\phi} \\ c_{+\xi} J_{n+1}(kr) e^{i(n+1)\phi} \\ c_{+\xi} J_{n+1}(kr) e^{i(n+1)\phi} \\ c_{-\xi} J_n(kr) e^{i(n+1)\phi} \end{array} \right) |\downarrow\rangle \right) + \left( \begin{array}{c} c_{+\xi} J_n(kr) e^{i\phi} \\ c_{-\xi} J_{n+1}(kr) e^{i(n+1)\phi} \\ c_{-\xi} J_{n+1}(kr) e^{i(n+1)\phi} \\ c_{+\xi} J_n(kr) e^{i(n+1)\phi} \end{array} \right) |\uparrow\rangle$$

with \(c_{\pm}\) as defined earlier and \(J_n(x)\) is a Bessel function. We analyze here weak scatterers, where the elastic scattering rate scales as \(\tau^{-1} \sim E_F\). Strong and resonant Coulomb scatterers [21] induces a similar change in the spin orientation. We approximate the potential by a step function, \(v(r) = V_0 [1 - \Theta(r - R)]\). The wave function inside the potential well is a superposition of two radial waves, finite at the origin and with different spin orientations. In graphene, Rashba spin coupling entangles spin and pseudospin and the change of spin in a scattering event depends on the evolution of the pseudospin. Unlike conventional semiconductors [2], the spins in different angular momentum channels differ, complicating the definition of the amount of spin relaxation in a scattering event. From the given incoming wave \(\Psi_{i\xi}^\pm\) with spin parallel to the momentum \(\vec{k}\) and incident angle \(\theta\), there are two possible outgoing waves \(\Psi_{el}^{\pm} (r, -\theta)\), \(\Psi_{el}^{\pm} (r, \theta)\), which satisfy conservation of energy and momentum. These can be written in a similar way as Eq.(2), where \(\theta' \approx -\theta + \Delta \cot(\theta)/(v_F k)\). We then define

$$S = \frac{\sum_n \left( r_n r_0 n + r_n' r_0' n \right) - \sum_n \left( r_0^2 n + (r_0' n)^2 \right)}{\sum_n (r_0^2 n + (r_0' n)^2)} \equiv \frac{\sum_n \left( r_n r_0 n + r_n' r_0' n \right) - \sum_n \left( r_0^2 n + (r_0' n)^2 \right)}{\sum_n (r_0^2 n + (r_0' n)^2)} ,$$

where \(r_n(r_0)\) and \(r_n'(r_0')\) are the scattering amplitudes for a given angular momentum channel n with (without) SO coupling and for the two possible outgoing waves \(\Psi_{el}^{\pm} , \Psi_{el}^{\pm}\) respectively. SO coupling changes the wavevector for one of the reflected waves, \(k' \approx k - \Delta/v_F\). The leading reflection coefficient, \(r_{n=0}, in\), in the absence of SO coupling depends on wavevector as \(r_{0}(k) \sim V_0 k R^2/v_F^2[21]\), so that \(r_{n=0}(k') - r_{n-0}(k) \sim V_0 \Delta R^2/(v_F)^2 S \sim \Delta/(v_F k)\). \(S\) vanishes if the spin is conserved in the scattering event. If the changes induced by a finite \(\Delta\) are small, this quantity should be proportional to the change in spin orientation during the scattering process. The change in spin orientation at each collision is \(\Delta/(v_F k)\). The total change of the spin after \(N_{\text{coll}}\) collisions is of order \(\sqrt{N_{\text{coll}} \Delta/(v_F k)}\). Dephasing occurs when \(\sqrt{N_{\text{coll}} \Delta/(v_F k)} \sim 2\pi\) after a time \(\tau_{so} = \tau_{el} N_{\text{coll}}\), where \(\tau_{el}\) is the elastic scattering time, and the Elliot-Yafet spin relaxation time is \(\tau_{so}^{\text{EY}} \sim (v_F k)^2/\Delta^2 \times (l_{\text{el}}/v_F) \sim (v_F k)^2/\Delta^2 \times \tau_{el}\). The spin diffusion length is related to the spin relaxation time by \(\lambda_{so} = \sqrt{D_{so}}\), where \(D = v_F^2/2\) so \(\lambda_{so}^{\text{EY}} \sim l_{\text{el}}(v_F k)/\Delta\).

The D’yakonov-Perel’ mechanism.- Between scattering events, the Rashba SO coupling acts as an effective magnetic field in the plane, \(\hat{B}_0(\vec{\sigma}) = \Delta(\vec{\sigma} \times \hat{e}_z)/2\), on the
The spin dynamics is a result of spins precessing in a fluctuating in-plane magnetic field governed by the momentum. Elastic scattering randomizes the momentum and the associated magnetic field. Averaging over many collisions, the spin orientation becomes random after a time [9]

\[ \tau_{so}^{DP} \approx v_F l_{el}^{-1} / \Delta^2, \]  

where \( \Delta \) is the magnetic length, which vanishes when the gauge field is weak. Using Eq. (5) for the magnetic length, which is the energy \( \Delta = \sqrt{2|\mathbf{B}||B_\perp|} \), we can rewrite the Hamiltonian for one valley \( K \),

\[
\mathcal{H} = \begin{pmatrix}
0 & 0 & v_F \hat{P} & 0 \\
0 & 0 & -i \Delta & v_F \hat{P} \\
v_F \hat{P} & i \Delta & 0 & 0 \\
v_F \hat{P} & 0 & 0 & 0
\end{pmatrix},
\]

where \( \Delta = \sqrt{2|\mathbf{B}||B_\perp|} \) and we consider a homogenous magnetic field to illustrate the main effect. The wave functions are

\[
\Phi_{n,\uparrow,\downarrow} = \left( \begin{array}{c} \mp i \phi_{n-1} x - x_0 \\ \phi_n (x - x_0) \end{array} \right) e^{iky \hat{\uparrow}, \hat{\downarrow}},
\]

with eigenenergy \( \epsilon_n = \pm v_F \sqrt{|n|}/l_B \) in terms of solutions of a particle in an harmonic oscillator potential \( \phi_n(x - x_0) \). The two components of \( \Phi_{n,\uparrow,\downarrow} \) correspond to the amplitudes in the two sublattice and \( x_0 = k_B l_B \) is the Landau level guiding center.

The Rashba SO coupling induces an interaction between electrons with spin up in one sublattice and electrons with spin down in the other sublattice. Therefore it is convenient to express the Hamiltonian (6) in the basis \( \phi_{n-1} [0, 0] \uparrow \uparrow \), \( \phi_n [0, 1] \uparrow \uparrow \), \( \phi_n [1, 0] \uparrow \uparrow \), \( \phi_{n+1} [0, 1] \uparrow \uparrow \), \( \mathcal{H} = \mathcal{H} \sqrt{\mathbf{B}}/l_B \):

\[
\mathcal{H} = \begin{pmatrix}
0 & i \sqrt{|n|} & 0 & 0 \\
-i \sqrt{|n|} & 0 & -i \Delta & 0 \\
i \Delta & 0 & 0 & i \sqrt{2|n+1|} \\
0 & 0 & i \sqrt{2|n+1|} & 0
\end{pmatrix},
\]

where \( \Delta = \Delta l_B/(v_F \sqrt{\mathbf{B}}) \). The eigenenergies of \( \mathcal{H} \) are:

\[
\epsilon_n = \sigma + \Delta^2 + 2n + \sqrt{\left(1 + \Delta^2\right)^2 + 4n \Delta^2}/2,
\]

where \( \sigma = + (\sigma = -) \) denotes electron (hole) like excitations and \( s = + (s = -) \) denotes spin. Let us consider the expectation value of the out-of-plane spin polarization of these states, \( p_z \). For \( n = 0 \) there are three physical states of which one has polarization \( \langle \hbar \rangle/2 \) \((1 + \Delta^2)/(1 + \Delta^2)\) and two have polarizations \( -\langle \hbar \rangle/2 \) \((1 + \Delta^2)\). For all states where \( n \geq 1 \) the polarizations are

\[
p_z = s h/2 \left(1 + \Delta^2\right)^2 + 4n \Delta^2 \right)^{-1/2}.
\]

The spin polarization differs between the lowest and highest Landau levels and the transition roughly occurs when \( 4 \sqrt{n} \Delta \approx 1 \). Transport is governed by states at the Fermi energy \( \epsilon_n \approx v_F \sqrt{|n|}/l_B \rightarrow v_F k_F \) and the condition \( 4 \sqrt{n} \Delta \approx 1 \) can be rewritten in terms of a critical value for the magnetic length, which exactly agrees with our semiclassical estimate for \( l_B \) in Eq. (5). Equation (9) demonstrates that when \( l_B \ll l_B^{(c)} \) the states are fully out-of-plane polarized, but the out-of-plane polarization vanishes when the gauge field is weak \( l_B \gg l_B^{(c)} \). The energy splitting is of the order \( \Delta \) for weak gauge fields and reduced by a factor \( l_B^{(c)}/l_B^2 \) for stronger gauge fields.
This change in the polarization direction of the eigenstates with increasing gauge fields has consequences for the spin relaxation. When the gauge fields vanish, the effective magnetic field is in plane so that spins out of plane relax twice as fast as spins in plane. In the regime around $l_B = l_B^{(c)}$, the in-plane and out-of-plane components of the effective magnetic field are comparable and we expect the spin-relaxation anisotropy to be reduced and eventually exhibit the opposite behavior, spins in plane relax faster than spins out of plane.

Experimental consequences.- We use typical parameters for graphene, $h\nu_F = 5.3 \times 10^{-10} eV/m$ and the enhanced Rashba coupling for a ripple of radius $R = 100 nm$ is $\Delta R = 100 nm = 1.7 \times 10^{-5} eV$. The Fermi wavelength depends on the electron doping $n$, $\lambda_F = \sqrt{2\pi/n}$. In Ref. [3] $n \approx 3.6 \times 10^{10} m^{-2}$ so $\lambda_F \approx 13 nm$ and $l_0 \approx 36 nm$, but considerably larger mean free paths have been measured and should be expected in clean systems in the future.

Comparing the DP and EY relaxation mechanisms, $\tau_{so}^{EY} \approx (v_F k_F)^2 / \Delta^2 \tau_p$ and $\tau_{so}^{DP} \approx \tau_{p}^{-1} / \Delta^2$, we find $\tau_{so}^{EY} / \tau_{so}^{DP} \approx (v_F k_F)^2 \sim (k_F l_0)^2$. Typically $(k_F l_0) > 1$, e.g. in Ref. [3] $(k_F l_0)^2 \approx 300$, so the DP mechanism is much more important than the EY mechanism.

The experimental trend that the spin-relaxation length is proportional to the mean free path $l_B^{(c)}$ is encouraging since it suggests clean systems should have a very long spin-relaxation length. However, this is at odds with our results for intrinsic graphene where the spin-relaxation length only weakly depends on the mean free path. The good news is that the computed spin relaxation length is long, we find $l_{prec} = 2\pi h\nu_F / \Delta R = 190 \mu m$ and hence the DP spin relaxation length is $\lambda_{f}^{DP} = l_{prec} / (2\sqrt{2}) \sim 20 \mu m$. On the other hand, in Ref. [3] $\lambda_{f} \sim 1.3 - 2 \mu m$. Our theory neither quantitively nor qualitatively explains the experiments in Ref. [3]. Adatoms such as $\ddot{H}$ enhance the SO interaction could be responsible for the discrepancy since the mobility of the experimental samples is relatively low $\Delta l_B^{(c)}$.

Our theory applies to cleaner, intrinsic graphene, possibly with less adatoms, where we predict novel spin-relaxation anisotropy effects. We expect it is possible to obtain longer spin-relaxation lengths than in Ref. [3] $l_B^{(c)}$, but that one eventually will enter the intrinsic regime where the DP mechanism prevents a further enhancement. Furthermore, in intrinsic graphene, gauge fields due to ripples are important. From the parameters above, we find a threshold magnetic field of $l_B^{(c)} \approx 200 nm$. Surface corrugations give rise to effective magnetic lengths of the order $l_B \approx 100 nm$, so already at these electron densities gauge fields are important and reduce the spin relaxation with respect to DP mechanism. Lower electron densities, which are feasible, should enhance the effects of the gauge fields. In ultra clean systems, $l_0 > l_B$,
[23] A. H. Castro Neto and F. Guinea, Phys. Rev. Lett 103, 026804 (2009).