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 its reduced dimensionality, 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 [2–8]. 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 A_3B_5 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 τ_{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 $\lambda_{sf} \sim 2$ μm was measured at room tem-

perature and it was indicated that λ_{sf} is proportional to the elastic mean free path l_{el}, 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 mecha-
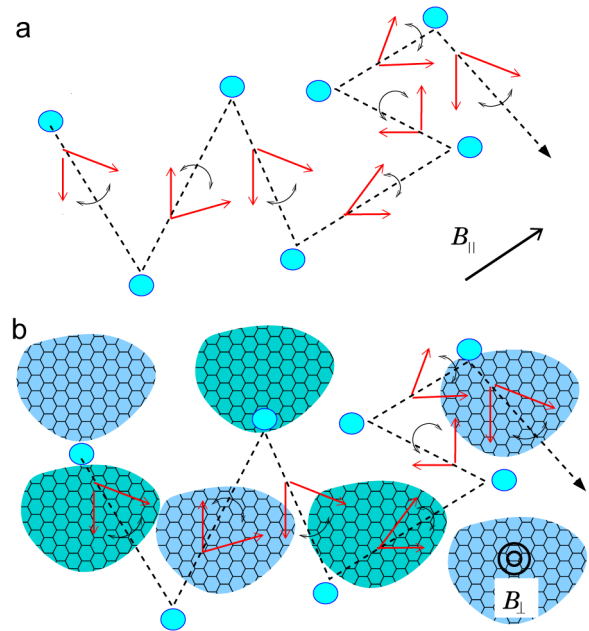

FIG. 1 (color online).   (a) Dyakonov-Perel spin relaxation: The SO coupling induces a momentum dependent effective field $\vec{B}_{\parallel}$ which changes direction randomly after scattering events, leading to spin relaxation. $\vec{B}_{\parallel}$ is "in plane" so spins directed perpendicular to the plane relax faster than as spins in the plane. (b) Gauge-field spin relaxation: Gauge fields due to ripples (light and gray blue areas) induce an effective field $\mathcal{B}_{\perp}$, which for a finite SO coupling leads to spin relaxation. $\vec{B}_{\perp}$ is "out of plane" so spins directed perpendicular to the plane relax slower than spins in the plane.

nisms, 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 mechanisms 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 ($\hbar = 1$)

$$\mathcal{H} = \pm v_F k (\vec{n} \cdot \hat{\sigma}) - \frac{\Delta}{2} (\hat{\sigma} \times \hat{s})_z, \qquad (1)$$

where $= +(-)$ corresponds to the $K(K')$ point, the spinor basis for K is $\Psi_K = (A_\uparrow, A_\downarrow, B_\uparrow, B_\downarrow)^T$ and for K' the components are reversed, $\Psi_{K'} = (B_\uparrow, B_\downarrow, A_\uparrow, A_\downarrow)^T$, A and B denote the lattice sites and $\uparrow$ and $\downarrow$ the electron spin. With this choice, the SO coupling is identical for K and K'. $\vec{n} = (\cos\theta, \sin\theta)$, $\tan\theta = k_y/k_x$, $k = \sqrt{k_x^2 + k_y^2}$ is the quasiparticle momentum with respect to the $K(K')$ corners of the hexagonal Brillouin zone, $\Delta = \Delta_{\text{curv}} + \Delta_\mathcal{E}$ [19] is the dominant Rashba SO coupling constant induced by curvature and/or 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

$$|\Psi_{k\xi s}\rangle = N_{\xi s} \left[s \begin{pmatrix} c_{-s} \\ c_s e^{i\theta} \end{pmatrix} i|\uparrow\rangle + \begin{pmatrix} c_s e^{i\theta} \\ c_{-s} e^{2i\theta} \end{pmatrix} |\downarrow\rangle \right] e^{i\vec{k}\cdot\vec{r}}$$

with energies $E_{k\xi s} = s\Delta_{\text{so}}/2 + \xi D$, $D = \sqrt{(v_F k)^2 + \Delta^2/4}$, where $s = \pm$ corresponds to the $\uparrow (\downarrow)$ spin states and $\xi = \pm$ denotes the pseudospin degeneracy, $c_{k\xi s} = \sqrt{E_{k\xi s}}/\sqrt{2D}$, $c_s \equiv c_{k\xi s}$ and $N_{\xi s} = \sqrt{2E_{k\xi s}}/(v_F k)$. These eigenstates are polarized in plane of the graphene layer. The spin precession length is $l_{\text{prec}} = 2\pi v_F/\Delta$.

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 discussed in [21]. Neglecting mixing of the K and K' valleys, an incoming wave with total angular momentum, $\mathcal{L} \equiv Ii\partial_\theta \pm \sigma_z/2 + s_z/2$, is an eigenstate of Eq. (1):

$$\Psi_{k\xi+}^{\text{in}}(r, \theta) \equiv \begin{pmatrix} c_- J_n(kr) e^{in\theta} \\ c_+ J_{n+1}(kr) e^{i(n+1)\theta} \end{pmatrix} i|\uparrow\rangle$$
$$+ \begin{pmatrix} c_+ J_{n+1}(kr) e^{i(n+1)\theta} \\ c_- J_{n+2}(kr) e^{i(n+2)\theta} \end{pmatrix} |\downarrow\rangle \qquad (2)$$

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 $\tau_{\text{el}}^{-1} \sim E_F$. Strong and resonant Coulomb scatterers [21] induce 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 [9], 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 Ψ^{in} with spin parallel to the momentum $\vec{k}$ and incident angle θ, there are two possible outgoing waves $\Psi_{k\xi+}^{\text{out}}(r, -\theta)$, $\Psi_{k\xi-}^{\text{out}}(r, \theta')$, which satisfy conservation of energy and momentum. These can be written in a similar way as Eq. (2), where $\theta' \approx -\theta + \Delta \cot(\theta)/(v_F k)$, respectively. We then define

$$\mathcal{S} = \frac{\sum_n (r_n r_{0n} + r'_n r'_{0n}) - \sum_n (r_{0n}^2 + (r'_{0n})^2)}{\sum_n (r_{0n}^2 + (r'_{0n})^2)}, \qquad (3)$$

where $r_n(r_{0n})$ and $r'_n(r'_{0n})$ are the scattering amplitudes for a given angular momentum channel n with (without) SO coupling and for the two possible outgoing waves Ψ_+^{out}, Ψ_-^{out} respectively. SO coupling changes the wave vector for one of the reflected waves, $k' \approx k - \Delta/v_F$. The leading reflection coefficient, $r_{n=0}$, in the absence of SO coupling depends on the wave vector as $r_0(k) \sim V_0 k R^2/v_F$ [21], so that $r_{n=0}(k') - r_{n=0}(k) \sim V_0 \Delta R^2/(v_F)^2$ and $\mathcal{S} \sim \Delta/(v_F k)$. $\mathcal{S}$ vanishes if the spin is conserved in the scattering event. If the changes induced by a finite Δ 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 $\Delta/(v_F k_F)$. The total change of the spin after N_{coll} collisions is of order $\sqrt{N_{\text{coll}}} \Delta (v_F k_F)^{-1}$. Dephasing occurs when $\sqrt{N_{\text{coll}}} \Delta (v_F k_F)^{-1} = 2\pi$ after a time $\tau_{\text{so}} = \tau_{\text{el}} N_{\text{coll}}$, where τ_{el} is the elastic scattering time, and the Elliot-Yafet spin-relaxation time is $\tau_{\text{so}}^{\text{EY}} \sim (v_F k_F)^2/\Delta^2 \times (l_{\text{el}}/v_F) \sim (v_F k_F)^2/\Delta^2 \times \tau_{\text{el}}$. The spin-diffusion length is related to the spin-relaxation time by $\lambda_{\text{so}} = \sqrt{D\tau_{\text{so}}}$, where $D = v_F^2 \tau/2$ so $\lambda_{\text{so}}^{\text{EY}} \sim l_{\text{el}}(v_F k_F)/\Delta$.

The Dyakonov-Perel mechanism.— Between scattering events, the Rashba SO coupling acts as an effective mag-

netic field in the plane, $\vec{B}_\parallel(\hat{\sigma}) = \Delta(\hat{\sigma} \times \hat{e}_z)/2$, on the spins [16,17]. 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]

$$\tau_{so}^{DP} \approx v_F l_{el}^{-1}/\Delta^2, \tag{4}$$

e.g., the spin-relaxation time is inversely proportional to the elastic scattering time. The DP spin-diffusion length is independent of the mean free path $\lambda_{so}^{DP} = v_F/(\sqrt{2}\Delta) = l_{prec}/(2\sqrt{2}\pi)$.

Effective gauge field.—Topological lattice defects, strains, and curvature change the hopping integrals between the lattice sites. These effects are captured by an induced effective gauge field $\mathcal{A}(\vec{r})$ which deflect electrons and change the electronic states at low energies [1,22]. $\mathcal{A}$ is related to the strain tensor $u_{ij}(\vec{r})$ describing topological lattice disorder, $\mathcal{A}_x \sim [u(\vec{r})_{xx} - u(\vec{r})_{yy}]$, $\mathcal{A}_y \sim u(\vec{r})_{xy}$ and give rise to a random out-of-plane magnetic field $\mathcal{B}_\perp = [\vec{\nabla} \times \mathcal{A}(\vec{r})]_z$ [22]. We will demonstrate that this out-of-plane orbital magnetic field together with the Rashba SO coupling can induce an out-of-plane spin polarization.

Let us first present a semiclassical argument for why the spin polarization changes from in plane to out of plane as the gauge field increases. $\mathcal{B}_\perp$ causes the electrons to move in cyclotron orbits with a radius $r_c = 2\epsilon/(ev\mathcal{B}_\perp)$, where v is the velocity and ϵ is the energy. A qualitative change in the electronic states occurs when the cyclotron orbit is smaller than the spin precession length, e.g., when $2\pi r_c = l_{prec}$. Using $v = v_F$, the graphene dispersion $\epsilon = v_F k_F$, and introducing the magnetic length $l_B = \sqrt{e/\mathcal{B}_\perp}$, there is a transition when the magnetic length is shorter than

$$l_B^c = \sqrt{l_{prec}\lambda_F}/(2\sqrt{2}\pi), \tag{5}$$

where $\lambda_F = 2\pi/k_F$ is the Fermi wavelength. In the low field regime, $l_B \gg l_B^c$, in-plane momentum is a good quantum number and via the SO interactions spins are polarized in plane. In contrast, in the strong field regime, $l_B \ll l_B^c$, out-of-plane angular momentum is a good quantum number, and the SO coupling changes the polarization of the states to out of plane.

We will now carry out a quantum mechanical calculation which will give more details and confirm our arguments above with the threshold value Eq. (5) for the gauge field. Inserting the gauge fields in the Hamiltonian in Eq. (1), results in a 4×4 Hamiltonian for one valley K,

$$\mathcal{H} = \begin{pmatrix} 0 & 0 & v_F\hat{\Pi} & 0 \\ 0 & 0 & -i\Delta & v_F\hat{\Pi} \\ v_F\hat{\Pi}^\dagger & i\Delta & 0 & 0 \\ 0 & v_F\hat{\Pi}^\dagger & 0 & 0 \end{pmatrix}, \tag{6}$$

where $\Pi = P_x - iP_y$, $P = -i\nabla + e\mathcal{A}$ and a similar Hamiltonian can be written for K', where the sign of the

gauge field is reversed. First, consider the solution of the Dirac equation when $\Delta = 0$. The results are gauge invariant so we choose the Landau gauge $\mathcal{A}_x = 0$, $\mathcal{A}_y = \mathcal{B}_\perp x$ and consider a homogenous magnetic field to illustrate the main effect. The wave functions are

$$\Phi_{n,\uparrow,\downarrow} = \begin{pmatrix} \mp i\phi_{n-1}(x - x_0) \\ \phi_n(x - x_0) \end{pmatrix} e^{iky}|\uparrow,\downarrow\rangle, \tag{7}$$

with eigenenergy $\epsilon_n = \pm v_F\sqrt{2|n|}/l_B$ in terms of solutions of a particle in a harmonic oscillator potential $\phi_n(x - x_0)$. The two components of $\Phi_{n,\uparrow,\downarrow}$ correspond to the amplitudes in the two graphene sublattices and $x_0 = kl_B^2$ 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 $\phi_{n-1}[1, 0]^T|\uparrow\rangle$, $\phi_n[0, 1]^T|\uparrow\rangle$, $\phi_n[1, 0]^T|\downarrow\rangle$, $\phi_{n+1}[0, 1]^T|\downarrow\rangle$, $\mathcal{H} = \tilde{\mathcal{H}}v_F\sqrt{2}/l_B$:

$$\tilde{\mathcal{H}} = \begin{pmatrix} 0 & i\sqrt{|n|} & 0 & 0 \\ -i\sqrt{|n|} & 0 & -i\tilde{\Delta} & 0 \\ 0 & i\tilde{\Delta} & 0 & i\sqrt{2|n + 1|} \\ 0 & 0 & -i\sqrt{|n + 1|} & 0 \end{pmatrix}, \tag{8}$$

where $\tilde{\Delta} = \Delta l_B/(v_F\sqrt{2})$. The eigenenergies of $\tilde{\mathcal{H}}$ are:

$$\tilde{\epsilon}_n^2 = \sigma[1 + \tilde{\Delta}^2 + 2n + s\sqrt{(1 + \tilde{\Delta}^2)^2 + 4n\tilde{\Delta}^2}]/2,$$

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 $(\hbar/2)(1 - \tilde{\Delta}^2)(1 + \tilde{\Delta}^2)$ and two have polarizations $-(\hbar/2)/(1 + \tilde{\Delta}^2)$. For all states where $n \geq 1$ the polarizations are

$$p_z = s\frac{\hbar}{2}[(1 + \tilde{\Delta}^2)^2 + 4n\tilde{\Delta}^2]^{-1/2}. \tag{9}$$

The spin polarization differs between the lowest and highest Landau levels and the transition roughly occurs when $4\sqrt{n}\tilde{\Delta} \approx 1$. Transport is governed by states at the Fermi energy $\epsilon_n \approx v_F\sqrt{2|n|}/l_B \rightarrow v_F k_F$ and the condition $4\sqrt{n}\tilde{\Delta} \approx 1$ can be rewritten in terms of a critical value for the magnetic length, which *exactly* agrees with our semiclassical estimate for l_B^c 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 Δ 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, $\hbar v_F = 5.3 \times 10^{-10}$ eV m and the enhanced Rashba coupling for a ripple of radius $R = 100$ nm is $\Delta_{R=100 \text{ nm}} = 1.7 \times 10^{-5}$ eV [19]. The Fermi wavelength depends on the electron doping n, $\lambda_F = \sqrt{2\pi/n}$. In Ref. [3] $n \approx 3.6 \times 10^{16}$ m^{-2} so $\lambda_F \approx 13$ nm and $l_{\text{el}} = 36$ 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, $\tau_{\text{so}}^{\text{EY}} \approx (v_F k_F)^2/\Delta_{\text{so}}^2 \tau_p$ and $\tau_{\text{so}}^{\text{DP}} \approx \tau_p^{-1}/\Delta_{\text{so}}^2$, we find $\tau_{\text{so}}^{\text{EY}}/\tau_{\text{so}}^{\text{DP}} \approx \tau_p^2 (v_F k_F)^2 \sim (k_F l_{\text{el}})^2$. Typically $(k_F l_{\text{el}})^2 \gg 1$, e.g, in Ref. [3] $(k_F l_{\text{el}})^2 \approx 300$, so the DP mechanism is much more important that the EY mechanism.

The experimental trend that the spin-relaxation length is proportional to the mean free path [3,18] 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 $l_{\text{prec}} = 2\pi\hbar v_F/\Delta_{R=100 \text{ nm}} = 190$ μm and hence the DP spin-relaxation length is $\lambda_{\text{sf}}^{\text{DP}} = l_{\text{prec}}/(2\pi\sqrt{2}) \sim 20$ μm. On the other hand, in Ref. [3] $\lambda_{\text{sf}} \sim 1.3$–2 μm. Our theory neither quantitively nor qualitatively explains the experiments in Ref. [3]. Adatoms such as hydrogen that locally enhance the SO interaction could be responsible for the discrepancy since the mobility of the experimental samples is relatively low [23].

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 Refs. [3,18], 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 length of $l_B^{(c)} \approx 200$ nm. Surface corrugations give rise to effective magnetic lengths of the order $l_B \approx 100$ nm [22] so already at these electron densities gauge fields are important and reduce the spin relaxation with respect to the DP mechanism. Lower electron densities, which are feasible, should enhance the effects of the gauge fields. In ultra clean systems, $l_{\text{el}} > l_B$, we expect $\tau_{\text{so}}^{\text{DP}} \rightarrow v_F l_B^{-1}/\Delta^2$ and the spin-relaxation time saturates. We encourage experiments in clean graphene, which will further elucidate the interplay between SO coupling, momentum relaxation, and gauge fields.

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