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Phys. Rev. Lett. **108**, 206808 — Published 17 May 2012

DOI: 10.1103/PhysRevLett.108.206808

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The differences between spin relaxation in graphene and in other materials are discussed. For relaxation by scattering processes, the Elliot-Yafet mechanism, the relation between the spin and the momentum scattering times acquires a dependence on the carrier density, which is independent of the scattering mechanism and the relation between mobility and carrier concentration. This dependence puts severe restrictions on the origin of the spin relaxation in graphene. The density dependence of the spin relaxation allows us to distinguish between ordinary impurities and defects which modify locally the spin-orbit interaction.

PACS numbers:

Introduction. Graphene is considered as a potential material for spintronics devices due to the weak spin-orbit (SO) interaction[1–3] and long spin lifetimes[4]. One of the most intriguing features of spintronics in graphene is that the observed spin relaxation time is significantly shorter than the values estimated theoretically. A number of recent experiments[5–7] investigate which spin relaxation mechanism plays the major role both in single layer and in bilayer graphene. The D'yakonov-Perel[8] and the Elliot-Yafet[9,10] mechanisms are usually discussed in the context of graphene[11,12], whereas the effect of hyperfine interactions is neglected due to the near absence of nuclear magnetic moments, as well as the Bir-Aronov-Pikus mechanism, which is important for heavily p-doped semiconductors[13]. Experiments suggest that the main source of spin relaxation in single layer graphene is extrinsic, lending support to the Elliot-Yafet mechanism. Longer spin lifetimes have been reported in bilayer graphene than in single layer[5], when the SO coupling in the bilayer is expected to be somewhat stronger[14].

The Elliot-Yafet mechanism takes into account the change in the spin polarization of a Bloch electron due to scattering by impurities, lattice defects or phonons. The Elliot relation establishes a linear relation between the spin relaxation time and momentum scattering time:

$$\tau_s = \frac{\tau_p}{\alpha} \tag{1}$$

where α can be interpreted as the spin-flip probability during a momentum relaxation event. Elliot deduced this relation by using a perturbative approach. Due to the SO coupling, Bloch states with well-defined spin polarization are not longer eigenstates of the complete Hamiltonian. In the case of conventional metals with a center of symmetry, two degenerate states can be defined for each value of the momentum[9]:

$$\left[a_{\mathbf{k}}\left(\mathbf{r}\right)\left|\uparrow\right\rangle + b_{\mathbf{k}}\left(\mathbf{r}\right)\left|\downarrow\right\rangle\right] e^{i\mathbf{k}\cdot\mathbf{r}} \tag{2}$$

$$\left[\left(a_{-\mathbf{k}}\left(\mathbf{r}\right)\right)^{*}\left|\downarrow\right\rangle - \left(b_{-\mathbf{k}}\left(\mathbf{r}\right)\right)^{*}\left|\uparrow\right\rangle\right] e^{i\mathbf{k}\cdot\mathbf{r}} \tag{3}$$

where the coefficients a, b are lattice-periodic due to the discrete translation symmetry. These two states are connected by spatial inversion and time reversal symmetries and form a Kramers' doublet. These states can be identified with spin-up and spin-down states because typically $|b| \ll 1$. Since the SO interaction couples electronic states with opposite spin projections in different bands (in the case of graphene the SO interaction couples π and σ bands), perturbation theory gives $|b| \approx \Delta_{SO}/\Delta E$, where ΔE is the energy difference between the two bands involved. Usually, $\Delta_{SO} \ll \Delta E$, as in the case of graphene.

The spin flip amplitude during the scattering by an obstacle with no spin degrees of freedom itself can be computed using the Born approximation, leading to $\alpha \approx \left\langle |b|^2 \right\rangle$ where the symbol $\langle\rangle$ expresses an average over the Fermi surface. These arguments are quite general and do not depend on the nature of the scatterers. Realistic calculations can be done in some cases, for instance in the case of III-V semiconductors[15,16].

The relation (1) holds experimentally for most conventional metals[17]. As we discuss below, doped graphene is not an exception. However, unlike ordinary semiconductors, the nature of the effective SO coupling acting on the graphene π electrons, which are the relevant ones in what concerns to transport properties, and importantly, the vanishing gap between conduction and valence electrons, makes the ratio τ_s/τ_p to depend strongly on the number of carriers through the Fermi energy. This result holds for different kind of defects, as it discussed next. A wide variety of experiments[18] suggest a linear scaling between τ_p and τ_s, with independence of the carrier concentration. Our analysis shows that this behavior cannot attributed to the Elliot-Yafet mechanism, opening the door to other extrinsically induced spin relaxation mechanisms, such as a defects which modify locally the spin-orbit interaction[19,20].

The model. In graphene, the intrinsic SO coupling can be neglected in comparison to the Rashba-like coupling, generated by perturbations which break spatial inversion, such as electric fields and ripples. If the perturbation changes slowly over scales larger than the lattice spacing, we can neglect intervalley hybridization[21]. Then, the Hamiltonian of the problem reads:

$$\mathcal{H} = -i\hbar v_F \vec{\sigma} \cdot \nabla + \frac{\Delta}{2} \left(\vec{\sigma} \times \vec{s} \right)_z \tag{4}$$

The Rashba-like term breaks the spatial inversion symmetry, and two degenerate eigenstates cannot be defined for a given momentum $\mathbf{k}$. A decomposition as the one in Eqs. (2)-(3) cannot be done, and the above Elliot's approach is not applicable. Moreover, the Rashba-like term (note that it is proportional to $\mathbf{v}_\mathbf{k} \times \mathbf{s}$, where $\mathbf{v}_\mathbf{k}$ is the velocity operator around each valley) entangles spin and sublattice degrees of freedom, complicating the definition of the amount of spin relaxation in a scattering event. The Bloch eigenstates of (4) read:

$$\Psi_{\mathbf{k},\pm} = \left[\begin{pmatrix} 1 \\ \frac{\epsilon_{\mathbf{k}\pm}}{\hbar v_F |\mathbf{k}|} e^{i\theta_\mathbf{k}} \end{pmatrix} \otimes |\uparrow\rangle \pm \right. \\ \left. \pm i \begin{pmatrix} \frac{\epsilon_{\mathbf{k}\pm}}{\hbar v_F |\mathbf{k}|} e^{i\theta_\mathbf{k}} \\ e^{2i\theta_\mathbf{k}} \end{pmatrix} \otimes |\downarrow\rangle \right] e^{i\mathbf{k}\cdot\mathbf{r}} \tag{5}$$

where $\theta_\mathbf{k} = \arctan\left(k_y/k_x\right)$ and $\epsilon^e_{\mathbf{k},\pm} = \pm\frac{\Delta}{2} + \sqrt{\left(\hbar v_F |\mathbf{k}|\right)^2 + \left(\frac{\Delta}{2}\right)^2}$, where e denotes electrons. A similar expression can be defined for holes by changing the sign of the second term. In what follows, we restrict the discussion to electrons. As we see, a spin direction cannot be uniquely defined for all momenta. When we take $\Delta = 0$, eigenstates (5) are Bloch states with well-defined projection of spin over the direction of motion, that is, helicity ±. This is not strictly true when $\Delta \neq 0$, but in the spirit of the Elliot's approach, we can identify each of these eigenstates with chiral states ±. This is justified from the point of view of perturbation theory, since for carrier concentrations of interest we have $\Delta/\epsilon_F \ll 1$. Thus, the effect of the Rashba-like coupling can be interpreted as the energy splitting ($\sim \Delta$) of bands with opposite chirality.

Let's consider now scattering by a potential $U\left(\mathbf{r}\right)$ diagonal in sublattice and spin degrees of freedom in the Born approximation. We study scattering in the chiral channels discussed above instead of the spin-up and spin-down channels as in the case of the Elliot's approach. This restriction complicates the definition of the amount of spin relaxation. To illustrate this, it is useful to calculate the scattering amplitudes in these channels in the absence of SO. Assuming and incoming Bloch state with energy $\epsilon = \hbar v_F k$ and positive chirality, it is easy to see that in that case (see Supplementary Information):

$$f_+\left(\theta\right) = -\left(\hbar v_F\right)^{-1} \sqrt{\frac{k}{8\pi}} U_\mathbf{q} e^{-i\theta} \left(1 + \cos\theta\right)$$

$$f_-\left(\theta\right) = -\left(\hbar v_F\right)^{-1} \sqrt{\frac{k}{8\pi}} U_\mathbf{q} i e^{-i\theta} \sin\theta \tag{6}$$

where $U_\mathbf{q}$ is the Fourier transformation of the scattering potential evaluated at the transferred momentum $\mathbf{k}' - \mathbf{k}$, and θ is the angle between the outcoming $\mathbf{k}'$ and incoming $\mathbf{k}$ momentum (see Fig. 1). If we repeat

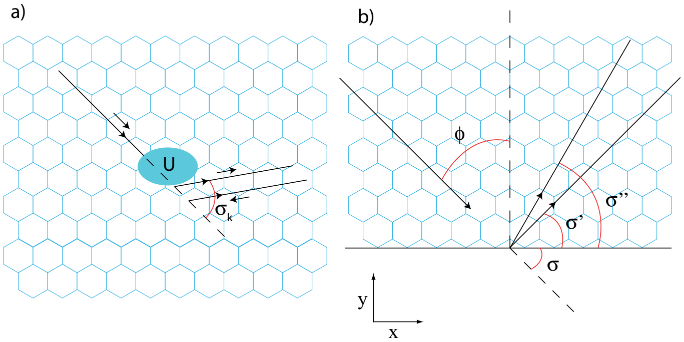

FIG. 1: a) Sketch of scattering by a potential $U\left(\mathbf{r}\right)$ in the chiral channels defined in the text. b) Sketch of scattering by a boundary.

the calculation in the spin-up and spin-down channels assuming an incoming state with spin up, then we obtain $f_\uparrow = -\left(\hbar v_F\right)^{-1} \sqrt{\frac{k}{8\pi}} U_\mathbf{q} \left(1 + e^{-i\theta}\right)$ and $f_\downarrow = 0$, since in the absence of SO there is no spin-flip. The scattering amplitude f_- is not zero in general (except for forward scattering) so that it cannot be related with a cross-section for a spin-flip process.

In order to study scattering in the chiral channels defined by the Rashba coupling we define the probability for a spin-flip process from the changes in the scattering in both chiral channels due to the presence of the SO coupling. We define the quantity:

$$S\left(\theta\right) = \frac{\sum_{\pm 1} \left| f^0_\pm\left(\theta\right) \right| \cdot \left| f^\Delta_\pm\left(\theta\right) - f^0_\pm\left(\theta\right) \right|}{\sum_{\pm 1} \left| f^0_\pm\left(\theta\right) \right|^2} \tag{7}$$

where the superscript Δ (0) indicates the presence (absence) of the Rashba-like coupling. This quantity vanishes when $\Delta = 0$, and it can be interpreted as a measure of the amount of spin relaxed in the direction defined by θ. As in the case of the Elliot's approach, the total amount of spin relaxation during a scattering event can be defined as the average of this quantity over the Fermi surface:

$$S = \langle S\left(\theta\right) \rangle = \frac{1}{2\pi} \int d\theta S\left(\theta, \epsilon = \epsilon_F\right) \tag{8}$$

In Ref. 11 the relation $S \sim \Delta/\epsilon_F$ was deduced in the case of weak scatterers. Here we show that this relation is general, and it does not depend on the nature of the scatterer, including strong scatterers or other impurity potentials which cannot be treated in the Born approximation, where the value of S cannot obtained from perturbation theory. The Born approximation suffices, however, to show how this behavior is implied by the nature of the SO coupling in graphene with independence of the precise scattering mechanism. It is not difficult to compute exactly $f^\Delta_\pm\left(\theta\right)$ (see Supplementary Information), but the picture provided by perturbation theory is enough to illustrate this behavior in the doped regime.

For an arbitrary scatterer, the value amplitude $f^0_\pm(\theta)$, as defined in (6), requires the use of non perturbative methods. The difference $f^\Delta_\pm(\theta) - f^0_\pm(\theta)$, however, can be obtained by expanding in powers of Δ/ϵ_F. This is easy to see in the Born approximation, where the substitution $\epsilon \to \epsilon \pm \Delta/2$ in expressions (6) has to be made in order to obtain $f^\Delta_\pm(\theta)$. An expansion in powers of Δ/ϵ is well defined, and it implies that $S(\theta) \sim \Delta/\epsilon$, independently of the scattering potential, $U_{\mathbf{q}}$, which factorizes in expressions (6). Assuming this behavior, the Elliot relation for graphene can be easily found. After N_{col} collisions, the change of spin polarization is of the order of $\sqrt{N_{col}}S$. Dephasing takes place after a time $\tau_s = N_{col}\tau_p$, when $\sqrt{N_{col}}S \sim 1$. Hence we obtain the relation:

$$\tau_s \approx \frac{\epsilon_F^2}{\Delta^2}\tau_p \tag{9}$$

This is the Elliot relation for graphene. As one can see, the ratio τ_s/τ_p depends on the carrier concentration through the Fermi energy. In what follows we compute exactly the amount of spin relaxation S for different kind of scatterers, generalizing the relation (9).

Results for different scatterers. The scattering amplitudes (6) can be calculated exactly in the presence of the Rashba-like coupling (see Supplementary Information). In the case of weak scatterers, we consider as scattering center a isotropic potential $U(\mathbf{r}) = V\vartheta(r-R)$, where $\vartheta(r-R)$ is a step function. In the case of Coulomb scatterers the scattering potential reads $U(\mathbf{r}) = -\hbar v_F \alpha/r$. Note that $|\alpha| < 1/2$, in other case the solutions of the Coulomb problem oscillate very fast and have no well-defined limit as $r \to 0$, which corresponds to the Dirac vacuum breakdown (the continuum description in terms of the Dirac Hamiltonian is not valid)[22]. The results are shown in Fig. 2. In both cases $S \sim \Delta/\epsilon_F$.

In order to study spin relaxation during scattering by a boundary we have to adapt the definition of S. We are going to consider a zig-zag termination for simplicity, since it defines the most general boundary conditions[23]. We consider as incoming wave a Bloch state $\Psi_{\mathbf{k},+}$ with energy ϵ, forming an angle ϕ with the direction perpendicular to the boundary. As it is deduced from Fig. 1, $\pi/2 + \sigma = \phi$, where $\sigma = \arctan(k_y/k_x)$. Two outgoing Bloch states exist satisfying conservation of energy and momentum in the direction parallel to the boundary. Then, the outgoing wave can be written as the superposition $\Psi_{out} = r_1\Psi_{\mathbf{k}_+,+} + r_2\Psi_{\mathbf{k}_-,-}$, where $\mathbf{k}_+$ ($\mathbf{k}_-$) forms an angle σ' (σ'') with the direction defined by the boundary (see Fig. 1), and $|\mathbf{k}_\pm| \equiv k_\pm = (\hbar v_F)^{-1}\sqrt{\epsilon^2 \mp \epsilon\Delta}$. We can define the amount of spin relaxed in the direction defined by ϕ as:

$$S(\phi) = \frac{|r_1^0| \cdot |r_1 - r_1^0| + |r_2^0| \cdot |r_2 - r_2^0|}{|r_1^0|^2 + |r_2^0|^2} \tag{10}$$

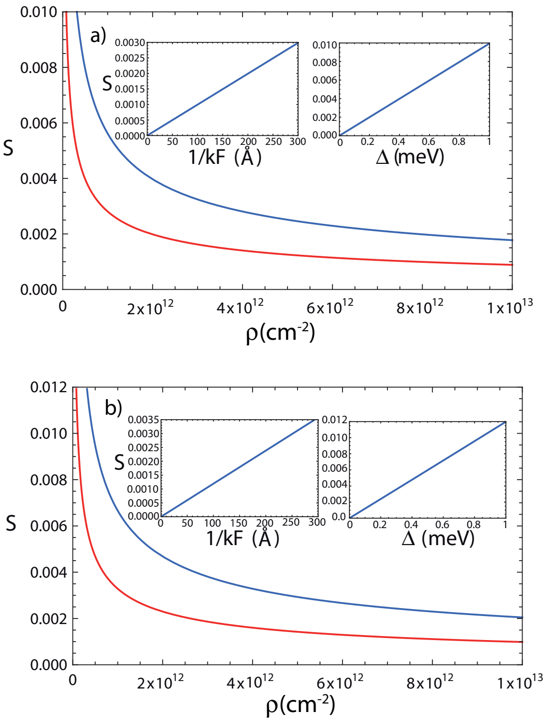

FIG. 2: S as a function of the carrier concentration for $\Delta = 1$ meV (in blue) and $\Delta = 0.5$ meV (in red). a) Weak scatterers ($R = 1$ Å and $V_0 = 0.1$ eV). b) Coulomb scatterers. These results are obtained by computing exactly the scattering amplitudes $f^\Delta_\pm$ and evaluating numerically Eq. (8). Insets: S for $\Delta = 0.1$ meV and $k_F = (\hbar v_F)^{-1}\epsilon_F = 0.01$ Å plotted as functions of k_F^{-1} and Δ respectively. A clear linear dependence is showed, as it is argued in the text.

where the superscript 0 refers to the reflection coefficients in the absence of the SO coupling. As before, the amount of spin relaxed by the boundary can be defined as the average, $S = \langle S(\phi)\rangle = \frac{1}{\pi}\int_{-\pi/2}^{\pi/2} d\phi S(\phi, \epsilon = \epsilon_F)$. By imposing zig-zag boundary conditions we obtain the following expressions for the reflection coefficients:

$$r_1 = -\frac{k_- e^{i\sigma} + k_+ e^{i\sigma''}}{k_- e^{i\sigma'} + k_+ e^{i\sigma''}} \tag{11}$$

$$r_2 = \frac{k_-\left(e^{i\sigma} - e^{i\sigma'}\right)}{k_- e^{i\sigma'} + k_+ e^{i\sigma''}} \tag{12}$$

Besides this, from conservation constrains we have $\sigma' = -\sigma$ and $\cos\sigma'' = \frac{k_+}{k_-}\cos\sigma$. To first order in the SO coupling, $\sigma'' = -\sigma - \frac{\Delta}{\epsilon}\cot\sigma + O\left(\Delta^2/\epsilon^2\right)$. Then, to first order in the SO coupling, the reflection coefficients in terms of the angle ϕ read:

$$r_1 = ie^{i\phi}\sin\phi + \frac{\Delta}{2\epsilon} + O\left(\frac{\Delta^2}{\epsilon^2}\right) \tag{13}$$

$$r_2 = -e^{i\phi}\cos\phi - \frac{\Delta}{2\epsilon} + O\left(\frac{\Delta^2}{\epsilon^2}\right) \tag{14}$$

The amount of spin relaxation can be estimated as $S =$

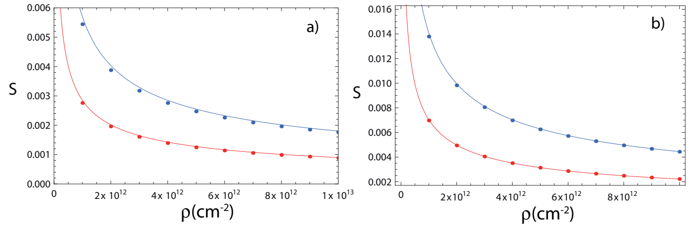

FIG. 3: S as a function of the carrier concentration for $\Delta = 1$ meV (in blue) and $\Delta = 0.5$ meV (in red). The dots correspond to the numerical evaluation of S, the continuum line to the analytical estimates of the text. a) Scattering by boundaries. b) Strong scatterers ($R = 1.4$ Å).

$\frac{2\Delta}{\pi\epsilon_F}$. This expression fits the exact result rather well, see Fig. 3.

In the case of strong scatterers, such as vacancies, the Born approximation fails. As in the case of boundaries, we need to extend the definition of S. Strong scatterers can be described as a circular void of radius of the order of the lattice constant. We can exploit the cylindrical symmetry of the problem by using the decomposition of the eigenstates of (4) into partial waves with well-defined generalized total angular momentum $J = l_z + \sigma_z/2 + s_z/2$, which is actually a global symmetry of the problem, where l_z is the third component of the orbital angular momentum operator $l_z = -i\left(x\partial_y - y\partial_x\right)$. For each incoming cylindrical wave with energy ϵ, there are two reflected waves with the same energy. A quantity analogous to Eq. (10) can be defined, considering scattering in all channels with well-defined J (see Supplementary Information). The amplitude for spin scattering can be calculated analytically $S \approx \frac{\pi\Delta}{2\epsilon_F}$ which fits very well the numerical evaluation of S, see Fig. 3.

The case of clusters of impurities[24] is studied within this formalism as well, since the Born approximation fails when the range of the scattering potential R is too large in such a way that the associated energy scale $\hbar v_F R^{-1}$ exceeds its strength, $VR \ll \hbar v_F$. The same behavior $S \sim \Delta/\epsilon_F$ is deduced (see Supplementary Information).

Discussion and conclusions. As we have seen, the averaged amount of spin relaxed during a scattering event behaves as $S \sim \Delta/\epsilon_F$, independently of the nature of the scatterer, implying the general relation $\tau_s \approx \epsilon_F^2 \tau_p/\Delta^2$. This unusual dependence on the carrier concentration ultimately arises from the absence of a energy gap between the conduction and valence π bands, and can be expected also in narrow gap semiconductors in the doped regime, where the Fermi energy is larger than the gap. In metals, spin relaxation is governed by intra-band transitions induced by the SO coupling at the Fermi energy, whereas in ordinary semiconductors it is dominated by inter-band transitions through the gap. In graphene we have a more complicated combination of both. This is clear when the Elliot-Yafet mechanism induced by the intrinsic SO coupling of graphene is studied, where the standard Elliot's approach is applicable (see Supplementary Information). The energy difference between the states at conduction and valence bands involved in the calculation is precisely $2\epsilon_F$, giving $\alpha \propto \Delta^2/\epsilon_F^2$. This result is not consistent with a linear scaling between the spin relaxation time and the diffusion coefficient at different gate voltages as it is observed in most of the experiments, suggesting that other mechanisms dominate spin scattering. It is interesting to consider in detail the experimental data of Ref. 18. The results show a sub-linear dependence of the diffusion constant on carrier density (proportional to the momentum scattering time) and also of the spin relaxation time. This is clearly inconsistent with our result if one assumes the Elliot-Yafet induced by defects in graphene as the main spin relaxation mechanism.

However, our theory explains recent experiments in CVD (Chemical Vapor Deposition) graphene-based spin valves[7]. In these experiments, the momentum scattering time shows a sub-linear dependence on carrier density, whereas the spin relaxation time shows a superlineal behavior, in agreement with our theoretical findings. This result implies that characteristic defects of CVD graphene samples, such as grain boundaries, are the main source of spin relaxation. In exfoliated samples, however, although there is a clear correlation between momentum scattering and spin relaxation, the deviation from our formula implies another mechanism (not strictly Elliot-Yafet) operating, essentially based on the scattering by defects which interact with the electron spins.

There are several candidates which could explain the experimental results in exfoliated graphene. The most relevant ones are: i) spin-flip induced by impurities covalently attached to graphene carbon atoms[19], ii) the effect of a fluctuating Rashba field created by ripples[25], iii) scattering by local spins. In the first two cases the spin-flip process is induced in regions where the SO coupling is locally enhanced, by heavy adatoms deposition[20], sp^3 hybridization due to resonant impurities[19], or ripples[25]. Importantly, a local enhancement of the SO coupling opens a new channel for spin relaxation apart from the one induced by momentum scattering (see Supplementary Information), as it is evident in recent works based on a kinetic spin Bloch equation approach[26,27]. Also in-plane strains could play a role, as it is pointed out in Ref. 11. This issue must be looked in more detail. In what concerns to the latter mechanism, the formation of local spins near defects as vacancies or resonant impurities is compatible with recent experimental[28] and theoretical[29] findings. This mechanism can be distinguished by the dependence of the spin lifetime on the injected current.

Acknowledgments: We appreciate useful discussions with R. Kawakami, B. J. van Wees, J. Fabian, V. I. Falko, B. Özyilmaz, S. Das Sarma, and A. K. Geim. AHCN acknowledges DOE grant DE-FG02-08ER46512 and ONR grant MURI N00014-09-1-1063. HO acknowledges financial support through grant JAE-Pre (CSIC, Spain). This work was also supported by MICINN (Spain) through grants FIS2008-00124 and CONSOLIDER CSD2007-00010.

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